A SIMPLE MODEL FOR THE ASSESSMENT OF INDOOR RADIONUCLIDE Pb-210 SURFACE CONTAMINATION DUE TO THE PRESENCE OF RADON

by

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The presented, very simplified model provides a possibility for estimation of surface Pb-210 activity, depending on the changes of Rn-222 concentration during the long-term radon presence inside the closed room. This can be useful for retrospective assessment of the average indoor radon concentration for certain historical period, based on the surface contamination by the radionuclide Pb-210 in a closed or poorly ventilated room over a long period of time. However, the surface Pb-210 contamination depends on the pattern of radon concentration changes, and in this model is supposed that the change of indoor radon concentration, which periodically enters the room, is affected only by the radioactive decay and the inserted amount of radon in each entry. So, each radon entry can be comprehended as a "net amount" of radon, or excess which remains inside the room due to radon's periodical in-out flow. It is shown, that under the conditions of the model, the achieved average value of radon concentration of 275 Bq/m^3, implies that the saturated surface contamination by the Pb-210 of 160 Bq/m^2 after approximately 150 years.

Key words: surface contamination, indoor radon concentration, Rn-222, Pb-210

INTRODUCTION

In the manuscript we considered an idealized example of a permanently closed room without ventilation, situated on the ground, where radon periodically enters in a certain amount. The bases for such assumption of radon periodical appearance are the published results [1], about measured radon variations (hourly, daily, and monthly) inside of a closed room due to the changes of temperature and the atmospheric pressure. However, the daily and seasonal changes of radon and radon progeny [2-4] may be significantly different for various locations, and even on the same location the pattern of changes can vary over time. The latter can be a reason for the discrepancy between the indoor radon concentration estimates based on the radon flux from soil and ground water, and measured values [5]. For the retrospective average radon concentration assessment [6], the Po-210 activity measurements can be performed [7, 8]. The complex experimental methods of determining the activity of implanted Po-210 and Pb-210 on the surface, such as a glass surface, are described in details in [9]. The indoor behaviour of radon and radon progeny, as well as their spatial distributions depend on several parameters [10, 11], including the presence of aerosols.

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For the simplicity, prompt radon periodical entries into the room ("pulses" of radon) are supposed in the model. As the result of radon decays and post-radon radio nuclides (Po-218, Pb-214, Bi-214, and Po-214), the radionuclide Pb-210 (half-life: 22.3 years) is produced (fig. 1).

In this model changes of radon concentration arise only from the radon radioactive decay and radon inserted by each periodic entry into the room through the floor from soil. On the first sight, one could realize that in the described model only "one direction" of periodical radon flow (from outside into the room) exists. However, that is just a suitable way to describe a situation where a certain "net amount" of radon remains inside the room as a consequence of radon's periodical in-out flow.

MODEL

The aim of the model is an estimation of room's surface contamination by the radionuclide Pb-210. The interior dimensions of the room observed in the model were 4 m 5 m 2.5 m (fig. 2) corresponding to the volume of 50 m^3 and the total area (walls, floor, ceiling) of 85 m^2. We analyzed the situation whereby at each radon entry the "injected" Rn-222 activity was 2500 Bq, or 50 Bq/m^3, keeping in mind the room volume.

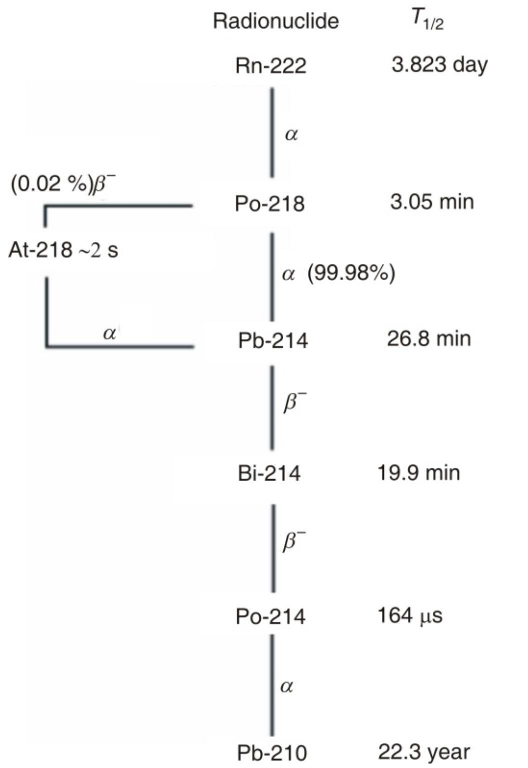

Figure 1. Decays of Rn-222 and post-radon radionuclides

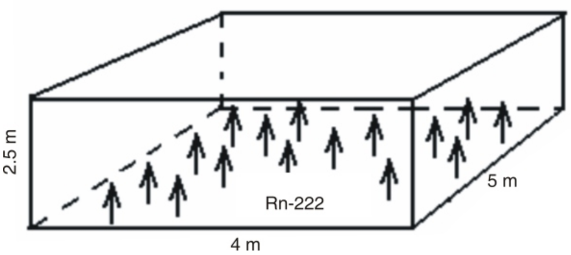

Figure 2. Dimensions of room

If at the initial moment (t = 0) N_0 nuclei of Rn-222 existed per unit volume of air (without progeny nuclei), then the time dependence of the Pb-210 activity per unit volume of air can be expressed as

$$a_{\mathrm{Pb}}(t) = N_0 (c_1 \mathrm{e}^{-\lambda_1 t} + c_2 \mathrm{e}^{-\lambda_2 t} + c_3 \mathrm{e}^{-\lambda_3 t} + c_4 \mathrm{e}^{-\lambda_4 t} + c_5 \mathrm{e}^{-\lambda_5 t} + c_6 \mathrm{e}^{-\lambda_6 t}) \quad (1)$$

In this expression, the decay constants $\lambda_1, \lambda_2, \lambda_3, \lambda_4, \lambda_5$, and λ_6 correspond to the Rn-222, Po-218, Pb-214, Bi-214, Po-214, and Pb-210, respectively, while the constants c_{m} (m = 1, 2,…, 6) are given as

$$c_m = \frac{\lambda_1 \lambda_2 \lambda_3 \lambda_4 \lambda_5 \lambda_6}{\prod_{\substack{i=1 \\ i \neq m}}^{6} (\lambda_i - \lambda_m)}$$

It is supposed that the Rn-222 appears every 24 hours (Δt = 24 h), fig. 3. The basis for this assumption lies in the fact that the similar diurnal variations of indoor radon concentration were often found by measurements [12]. As it can be seen, the Rn-222 activity per unit volume of air at the moment t represents the sum of activities resulting from the first, the second and the third radon appearance.

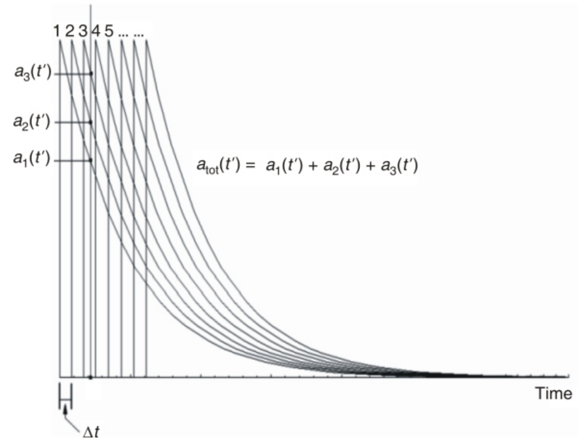

Figure 3. Periodical entries of radon into the room

Generally, the Rn-222 activity per unit volume of air at the moment of nth radon entry ($t = n\Delta t$, n = 0, 1, 2, 3, 4, 5, ...) can be expressed in the following way

$$\begin{aligned}
&a(t = 0) = \lambda_1 N_{IN}, \; n = 0 \\
&a(t = \Delta t) = \lambda_1 N_{IN} + \lambda_1 N_{IN} \mathrm{e}^{-\lambda_1 \Delta t}, \; n = 1 \\
&a(t = 2\Delta t) = \lambda_1 N_{IN} + \lambda_1 N_{IN} \mathrm{e}^{-\lambda_1 \Delta t} + \lambda_1 N_{IN} \mathrm{e}^{-\lambda_1 2\Delta t}, \\
&\quad n = 2 \\
&\vdots \\
&a(t = n\Delta t) = \lambda_1 N_{IN} (\mathrm{e}^{-\lambda_1 n\Delta t} + \mathrm{e}^{-\lambda_1 (n-1)\Delta t} + \mathrm{e}^{-\lambda_1 (n-2)\Delta t} \\
&\quad + \mathrm{e}^{-\lambda_1 (n-3)\Delta t} + \ldots + \mathrm{e}^{-\lambda_1 \Delta t} + 1) \quad (2)
\end{aligned}$$

Here N_{IN} represents the number of Rn-222 nuclei inserted into the room per unit volume by each radon entry, corresponding to an increase in activity concentration of 50 Bq/m^3 (N_{IN} = 2.3826543 10^7/m^3). This parameter value can be chosen in the model, as well as the number of radon entries and the time between the radon appearances. Precisely speaking, "nth entry" means nth entry of radon after the initial entry at the moment t = 0.

The activity per unit volume of air at the middle of the time interval between nth and (n + 1)th entry of radon ($t = n\Delta t + \Delta t/2$) is taken as a measure of Rn-222 concentration inside room. That can be found by multiplying the eq. (2) with $\mathrm{e}^{-\lambda_1 \Delta t/2}$, *i. e.*

$$\begin{aligned}
&a(t = n\Delta t + \Delta t/2) = \\
&\lambda_1 N_{IN} (\mathrm{e}^{-\lambda_1 n\Delta t} + \mathrm{e}^{-\lambda_1 (n-1)\Delta t} + \mathrm{e}^{-\lambda_1 (n-2)\Delta t} + \\
&\mathrm{e}^{-\lambda_1 (n-3)\Delta t} + \ldots + \mathrm{e}^{-\lambda_1 \Delta t} + 1)\mathrm{e}^{-\lambda_1 \Delta t/2} \quad (3)
\end{aligned}$$

By using of MATHEMATICA software, the time dependence of Rn-222 activity per unit volume is obtained for n = 200 radon entries, corresponding to a total time of 200 days (4800 h) from the moment when the room was closed (fig. 4).

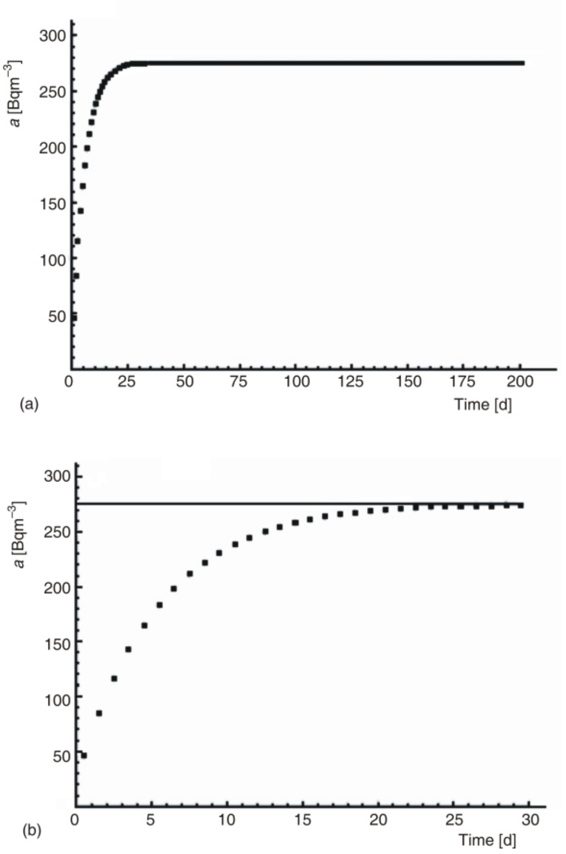

Figure 4. Time dependence of radon concentration in the room; (a) the radon concentration during 200 days; (b) the radon concentration during first 30 days

It can be seen that the saturation value of radon concentration of 275 Bq/m^3 is achieved, which is related, as mentioned before, to the middle of time interval between two successive radon appearances. This saturation value (*i. e.* average value) is practically achieved during first 30 days, fig. 4(b). According to the previously defined conditions in the model (amount of inserted radon by each entry, time interval between two entries), the radon concentration will, in fact, vary nearly within the boundaries $a\ (t = n\Delta t + \Delta t/2) \pm 25$ Bq/m^3, fig. 5.

The increase of Pb-210 activity per unit volume of air is afterwards analyzed in the model. The number of Rn-222 entries was $n = 5 \cdot 10^5$, corresponding to the total time of $5 \cdot 10^5$ days (1370 years). In this phase of the model it is supposed that the resulting activity of Pb-210 existed only in the air. According to eq. (1), the Pb-210 concentration (expressed in [Bqm^{-3}]) at the moment of n-th radon entry ($t = n\Delta t$) can be found as

$$a_{Pb}(t = n\Delta t) = N_{IN}\sum_{k=1}^{n}(c_1 e^{-\lambda_1 k\Delta t} + c_2 e^{-\lambda_2 k\Delta t} + c_3 e^{-\lambda_3 k\Delta t} + c_4 e^{-\lambda_4 k\Delta t} + c_5 e^{-\lambda_5 k\Delta t} + c_6 e^{-\lambda_6 k\Delta t}) \quad (4)$$

where the inserted number of radon nuclei by each radon entry, N_{IN}, per unit volume of air corresponds to the increase of radon concentration of 50 Bq/m^3.

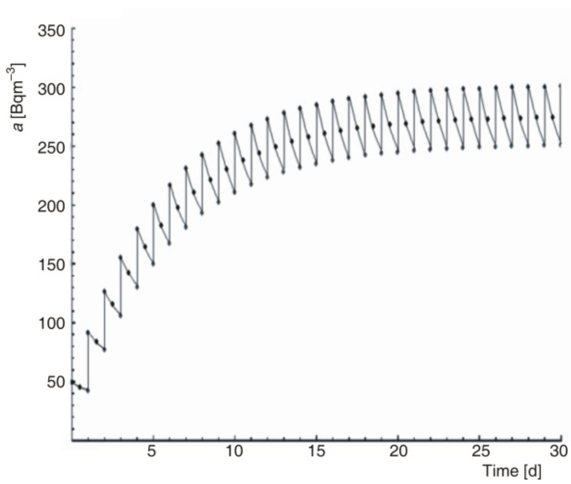

Figure 5. Variation of radon concentration

The concentration of the Pb-210 and its total activity $A^{Pb}_{tot}(t) = a_{Pb}(t)V$, depending on the time, are given in tab. 1, in the third and the forth column, respectively. In the next step of model it is supposed that the total activity of Pb-210, existing in the air at the certain moment, will be uniformly deposited on the interior surface of the room (the influence of aerosols on spatial distribution of radon progeny is neglected in the model). Consequently, the surface Pb-210 contamination is $a^{Pb}_S(t) = A^{Pb}_{tot}/S$, where S is the total area of room interior (tab. 1, last column).

The time dependence of surface Pb-210 contamination is presented in fig. 6. The graph represents the function $a^{Pb}_S(t) = \alpha + \beta(1 - e^{-\gamma t})$ obtained by fitting of results from tab. 1, $\alpha = -0.068023638$, $\beta = 162.09468$, and $\gamma = 0.031123624$.

The saturation of Pb-210 surface activity is achieved after approximately 150 years.

In a similar way we obtained the set of curves which represent the time dependence of Pb-210 surface contamination for different average Rn-222 concentrations, fig. 7.

The presented curves correspond to the following average radon concentrations: 50, 100, 200, 300, 400, 600, 800, and 1000 Bq/m^3.

These curves can be used for the retrospective assessments of average Rn-222 concentrations. For example, if the measured Pb-210 contamination of appropriate surface (such as glass) in a closed, or poorly ventilated room was found to be 40 Bq/m^2, and knowing the approximate time interval during which the room was closed (for example, about 6 years), it is possible to estimate the average Rn-222 concentration of about 400 Bq/m^3 for this long time interval using data from fig. 7. Also, for other possible combinations of measured surface Pb-210 contaminations and the investigated time intervals, the average radon concentrations can be found from fig. 7.

Table 1. The surface Pb-210 contamination

Number of radon entries into room	Time (t) elapsed from the moment of first radon entry [years]	Expected activity of Pb-210 per unit volume of air [Bqm^{-3}]	The total activity of Pb-210 inside of room [Bq]	The activity of Pb-210 per unit of radon activity inside of room	The surface contamination, $a_S^{Pb}(t)$, due to uniform deposition of Pb-210 from the air [Bqm^{-2}]
5	0.0137	0.0463	2.32	0.00028	0.027
10	0.0274	0.135	6.75	0.00057	0.079
50	0.137	1.053	52.6	0.00383	0.62
100	0.27	2.22	111	0.0081	1.31
200	0.55	4.54	227	0.0165	2.67
300	0.82	6.84	342	0.0249	4.02
400	1.10	9.12	456	0.0332	5.36
500	1.37	11.38	569	0.0414	6.69
600	1.64	13.62	681	0.0495	8.01
700	1.92	15.84	792	0.0576	9.32
800	2.19	18.05	902	0.0656	10.61
900	2.46	20.23	1011	0.0735	11.89
1000	2.74	22.40	1120	0.0815	13.18
5000	13.70	95.53	4776	0.347	56.2
10000	27.40	158.0	7900	0.575	92.9
50000	137.0	271.9	13595	0.989	160.0
500000	1370.9	275	13750	1.000	161.8

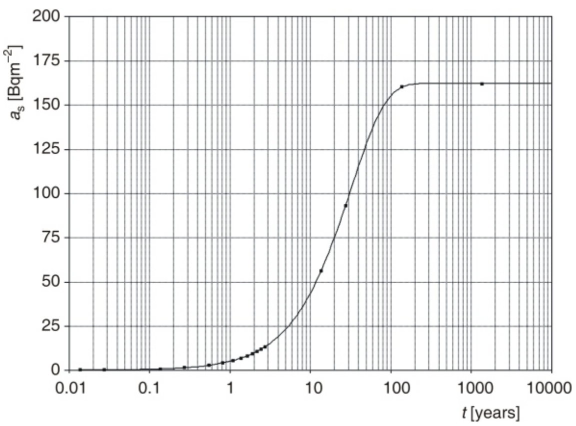

Figure 6. Increase of surface Pb-210 contamination for Rn-222 average concentration of 275 Bq/m^3

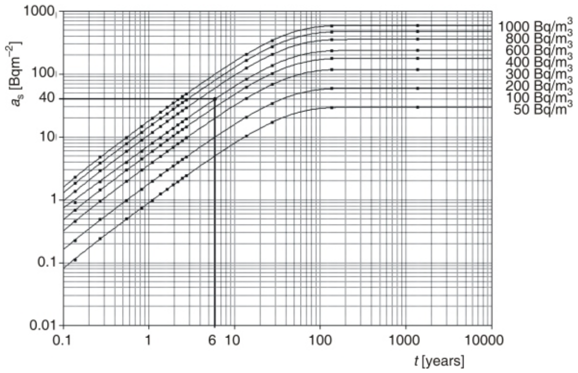

Figure 7. The expected surface Pb-210 contamination over time for different average Rn-222 concentrations. The presented curves correspond to the following average radon concentrations: 50, 100, 200, 300, 400, 600, 800, and 1000 Bq/m^3

CONCLUSIONS

Although very simplified, the presented model enables estimation of Pb-210 indoor surface contamination under specific conditions, such as permanently closed room, where only periodical radon variations occur. The model provides flexibility in changing of parameters such as the period between two successive radon appearances and the amount of radon excess per each radon entry.

This model can be useful for a retrospective assessment on average indoor radon concentration inside the closed objects which were virtually without ventilation for a very long time, on the basis of Pb-210 surface contamination data. However, the limitations caused by the simplifications assumed in the model, and by the supposed pattern of radon changes should be had in mind. For a more precise model, the influence of a selected radon variations pattern on Pb-210 surface contamination should be further tested.

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AUTHOR CONTRIBUTIONS

Theoretical analysis was carried out by D. S. Mrdja and I. S. Bikit, while additional calculations were performed by M. J. Vesković, S. M. Forkapić, N. M. Todorović, J. B. Nikolov, and K. I. Bikit. The manuscript was written and the figures were prepared by all the authors.

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ЈЕДНОСТАВАН МОДЕЛ ЗА ПРОЦЕНУ ПОВРШИНСКЕ КОНТАМИНАЦИЈЕ ЗАТВОРЕНОГ ПРОСТОРА РАДИОНУКЛИДОМ Pb-210 УСЛЕД ПРИСУСТВА РАДОНА

Приказан је врло поједностављен модел који омогућава процену површинске активности радионуклида Pb-210 у зависности од промена концентрације Rn-222, током дугорочног присуства радона унутар затворене просторије. Ово може да буде корисно за ретроспективну процену средње концентрације радона за одређени временски интервал, уколико се утврди вредност површинске контаминације радионуклидом Pb-210, за просторију која је била затворена или слабо вентилирана током дугог периода времена.

Површинска Pb-210 контаминација зависи од начина на који је концентрација радона током времена варирала. У овом раду је претпостављено да на промену концентрације радона, који периодично улази у дату просторију, утиче само радиоактивни распад и унесена количина радона при сваком уласку. Сваки улазак радона овде се може схватити као "нето износ" радона или вишак, који преостаје унутар просторије услед периодичног тока радона унутар-напоље. Показано је да, под условима овог модела, достигнута средња вредност концентрације радона од 275 Bq/m^3, имплицира сатурациону површинску контаминацију радионуклидом Pb-210 од 160 Bq/m^2 након приближно 150 година.

Кључне речи: површинска контаминација, концентрација радона у затвореном простору, Rn-222, Pb-210