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ORIGINAL PAPER

Dynamics of radon and its decay products inside an accumulative chamber

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Abstract. The calculations of radon and its decay products accumulation dynamics inside a chamber designed for measurements of radon flux density from the soil surface were carried out in the present work. Dynamics of alpha-, beta-particles and gamma-ray yields under decay of accumulated radionuclide nuclei was calculated based on the accumulated activity. Ion production rate inside the accumulative chamber due to ionizing radiation of different types was estimated. Thoron and its decay products accumulation regularities inside the accumulative chamber were also estimated and potential effect of thoron on measurement results (if counting regime is used) was analyzed in detail. Dynamic equations of the studied radiation characteristics, their solutions and obtained results analysis are presented.

Key words: radon • thoron • accumulative chamber • ionizing radiation yield • ion production rate

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Introduction

The background of this work was a need to search for a simple, cheap and reliable method of radon flux density measurement using an accumulative chamber. The method was intended to be realized in a measuring complex able to operate safely in the acute continental climate without human intervention over a long period using an independent supply source for applying at the monitoring stations. As the ultimate goal is to obtain, a continuous informative time series of radon field data, sample data high frequency are required. Satisfactory complexes for continuous measurement of radon flux density from the ground surface can be useful in studying connection between subsoil radon field dynamics and earth crust deflected mode changing, and lithosphere-atmosphere bonds as well. Their use at the monitoring stations together with measurement devices for radon pore activity in the soil air will allow to increase the predictive estimate reliability [26], and will enable to determine and specify radon transport model parameters in geological medium and ground atmosphere.

The general principle used in the existing devices is the following: radon entering from soil into an accumulation chamber gets either immediately into the measuring chamber or after finishing of accumulating time. Accumulated radon activity is measured by different methods and devices. Radon detecting is mainly performed by alpha radiation of radon and/or its short-lived decay products (^{218}Po and ^{214}Po) (scintillation [1, 5, 8, 20, 21] and semiconductor [7, 25] methods). In current

and pulse ionization chambers [6, 9, 13, 17] and electrets ion chambers [14] radon detecting is performed by air ionization produced mainly by alpha particles. Using activated carbon collectors for radon accumulation, radon activity is determined by gamma emission of short-lived radon decay products (^{214}Pb and ^{214}Bi) [16, 19, 22, 23]. Accumulated activity is measured using an solid-state nuclear track detector (SSNTD) technique as well [2, 15]. The way of accumulated radon measurement by beta emission of short-lived radon decay products is less spread in world practice (scintillation method [4] and ionization method [28]).

The objectives of this paper are the following: simulation of dynamics of radon activity and its decay products inside the accumulative chamber meant for measurement of radon flux density from the ground surface; calculation of alpha-, beta-particles and gamma-rays yield and ion production rate produced by them. Based on calculations, the following can be performed: the choice of more informative parameters measured in an accumulative chamber for further radon flux density evaluation under the desired conditions of experiment.

Calculation of the activity dynamics of radon and its decay products inside the chamber

The system of equation describing the balance of radionuclide activity (radon and its short-lived decay products) inside the accumulative chamber can be written as follows, considering only the processes of radionuclide formation and decay:

$$\begin{aligned}
\frac{\partial A_1(t)}{\partial t} &= K - \lambda_1 A_1(t) \\
\frac{\partial A_2(t)}{\partial t} &= \lambda_2 A_1(t) - \lambda_2 A_2(t) \\
\frac{\partial A_3(t)}{\partial t} &= \lambda_3 A_2(t) - \lambda_3 A_3(t) \\
\frac{\partial A_4(t)}{\partial t} &= \lambda_4 A_3(t) - \lambda_4 A_4(t) \\
A_5(t) &= A_4(t)
\end{aligned} \qquad (1)$$

where: A_i (t) is the volumetric activity of i-radionuclide (Bq·m^{-3}); an index i = 1,2,3,4,5 is correspondingly for ^{222}Rn, ^{218}Po (RaA), ^{214}Pb (RaB), ^{214}Bi (RaC) and ^{214}Po (RaC'); λ_i is the i-radionuclide decay constant (s^{-1}); $K = q \cdot S/V$ is the intensity of soil radon entry from the surface area S (m^2), into the accumulative chamber (Bq·m^{-3}·s^{-1}); V is the volume of the chamber (m^3); q is the radon flux density from the ground surface (Bq·m^{-2}·s^{-1}).

Sedimentation rate (sedimentation by gravity) of free decay daughter products is rather low, less than 6.7×10^{-8} m·s^{-1} [10], therefore it can be neglected. Deposition rates (by Brownian diffusion) on the surface of chamber and measuring devices are not considered in Eq. (1). However, according to the data [27], the deposition rates of free ^{218}Po and ^{214}Pb onto solid surfaces have a similar changing range from 0.02 to 0.09 cm·s^{-1} and, on the average, they are equal to 0.05 cm·s^{-1}, so it is necessary to take them into account for more accurate calculation.

Solving the equation system (1) let us assume that the value of radon flux density is constant during the accumulation time (~ 1 h). Formulating an equation system (1), we guess that the closed chamber is hermetic and radon leakage, back-diffusion processes are absent. Leakage and back-diffusion effects are better to investigate in practice with definite device [25].

The solutions of equations system (1) are as follows:

$$\begin{aligned}
A_1(t) &= C_1 e^{-\lambda_1 t} + K/\lambda_1 \\
A_2(t) &= C_2 e^{-\lambda_2 t} + \frac{C_1\lambda_2}{\lambda_2 - \lambda_1} e^{-\lambda_1 t} + \frac{K}{\lambda_1} \\
A_3(t) &= C_3 e^{-\lambda_3 t} + \frac{C_2\lambda_3}{\lambda_3 - \lambda_2} e^{-\lambda_2 t} + \frac{C_1\lambda_3\lambda_2}{(\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)} e^{-\lambda_1 t} \\
&\quad + \frac{K}{\lambda_1} \\
A_4(t) &= C_4 e^{-\lambda_4 t} + \frac{C_3\lambda_4}{\lambda_4 - \lambda_3} e^{-\lambda_3 t} + \frac{C_2\lambda_4\lambda_3}{(\lambda_4 - \lambda_2)(\lambda_3 - \lambda_2)} e^{-\lambda_2 t} \\
&\quad + \frac{C_1\lambda_4\lambda_3\lambda_2}{(\lambda_4 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)} e^{-\lambda_1 t} + \frac{K}{\lambda_1}
\end{aligned} \qquad (2)$$

Equations coefficients C_i are evaluated based on initial conditions $A_i(0) = 0$.

Figure 1 represents the accumulation dynamics of radon and its decay products in the accumulative chamber with a constant value of radon flux density from the soil surface, assumed in calculations as 10 mBq·m^{-2}·s^{-1} (a typical value observed for loams on the assumption of radon transport carried out via diffusion only). The following parameters have been selected for calculations: chamber volume – 3 dm^3, height – 0.1 m, accumulation time – 1 h. One hour accumulation time of radon has been chosen based on optimization conditions. Limitation of a range of upper boundary is caused by the requirement of higher frequency discretization of time series of radon field data.

Figure 1 shows that in an hour radon activity inside the chamber will reach 350 Bq·m^{-3} at a given radon flux density. As radon flux density from the soil surface is not only caused by diffusion, but by advection as well, the flux value can be significantly greater in the real situation. Thus, we can assume that calculating curves represent the lower limit of possible range of values changing of accumulated activities inside the chamber.

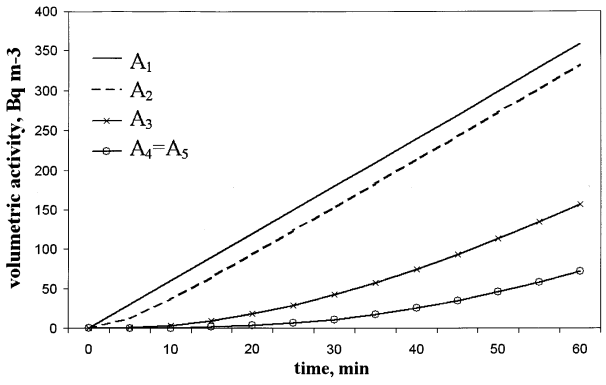

Fig. 1. Volumetric activity dynamics of radon and its decay products.

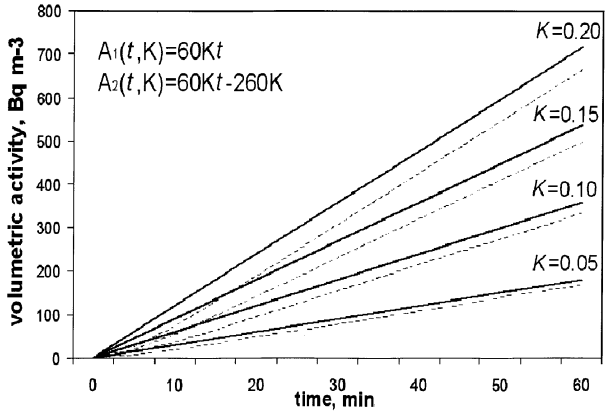

Fig. 2. Increase of volumetric activity of ^{222}Rn (solid line) and ^{218}Po (dotted line) with time at different values of intensity K.

As follows from Fig. 1, radon and ^{218}Po activities increase linearly (or practically linearly) over time, so curves $A_1(t)$ and $A_2(t)$ can be approximated by the linear function $A(t) = at + b$, where coefficients a, b will vary depending on intensity value of soil radon entry into the accumulative chamber (K).

Figure 2 represents the ^{222}Rn and ^{218}Po volumetric activity dynamics at different intensity values K, fitting radon flux density from 5 to 20 mBq·m^{-2}·s^{-1}. Approximation functions for ^{222}Rn and ^{218}Po activity in Bq·m^{-3} depending on K in Bq·m^{-3}·s^{-1} and accumulation time, t (in min) are also given. The calculations showed that coefficients a in linear functions of approximation for ^{222}Rn and ^{218}Po are equal. The revealed regularities can be used in method development of radon flux density measurement or in interpretation of radon accumulation curves inside the accumulative chamber obtained experimentally. Calculations have shown that in an hour ^{218}Po activity reaches 90%, ^{214}Pb – 40%, ^{214}Bi and ^{214}Po only 20% of radon activity.

Calculation of ionizing particles yield in the accumulative chamber

For the indicated volumetric activity values, the yield χ_j of j-type ionizing radiation formed by radioactive decay of radon and its decay products in the accumulative chamber in one second was evaluated

$$\chi_j(t) = V \cdot \sum_i (X_{ij} \cdot A_i(t)) \quad (3)$$

where: X_{ij} is the yield of j-type ionizing radiation per one nuclear decay of i-radionuclide.

Values of X_{ij} were taken from [12]. Alpha-particle yield dynamics $\chi_\alpha(t)$ is caused by nuclei ^{222}Rn, ^{218}Po and ^{214}Po accumulation inside the accumulative chamber, for beta-particles $\chi_\beta(t)$ and gamma-rays $\chi_\gamma(t)$ – by nuclei ^{214}Pb and ^{214}Bi accumulation.

Considering the possibility for particle yield and its dynamics detected by different nuclear radiation detectors, for further translation into radon flux density value, let us calculate ionizing radiation yield $\chi_j(t_2 - t_1)$ of j-type in an accumulative chamber for a period of time $(t_2 - t_1)$ based on the following relation

$$\chi_j(t_2 - t_1) = V \cdot \sum_i (X_{ij} \cdot N_i(t_2 - t_1)) \quad (4)$$

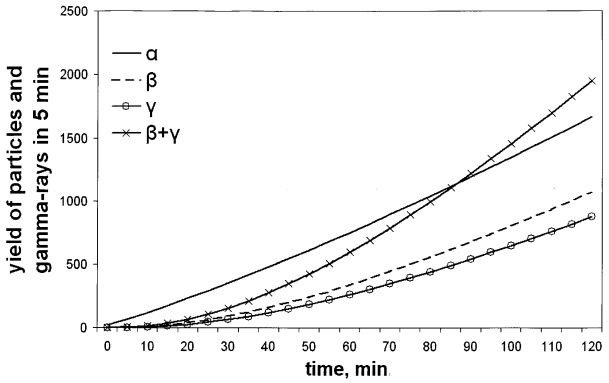

Fig. 3. Dynamics of ionizing particles and gamma-rays quantity formed in 5 min.

where $N_i(t_2 - t_1)$ is the number of i-type radionuclide decays for period of time $(t_2 - t_1)$ in 1 m^3 calculated by the formula

$$N_i(t_2 - t_1) = \int_{t_1}^{t_2} A_i(t)dt \quad (5)$$

Figure 3 represents the changing of ionizing particles and gamma rays formed in 5 min (time of one measurement) in the accumulative chamber with a volume of 3 dm^3. The time of one measurement depends on the chosen device characteristics and required statistics. For example, the measurement time of 5 min was chosen for our device [28].

Based on dynamics, represented in Fig. 3, we can choose a detector and radiation type for radon detection. Alpha-particle flux registered by a detector is by two times greater than beta and gamma radiation fluxes under the same measurements conditions. However, using gas-discharge counters detecting total beta and gamma radiation, we can get a detector response comparable to a signal from alpha radiation by providing similar measurement geometry and detector efficiency. As follows from Fig. 3, curves α and β + γ are crossed in the accumulative period of 90 min.

Calculation of ion production rate inside the accumulative chamber

Next, to determine the fitness for the use of this parameter in measurement of radon flux density, the calculations of ion production rate dynamics by different types of ionizing radiation inside the accumulative chamber were carried out.

Alpha-particles ranges for radon, ^{218}Po and ^{214}Po in the air are equal to 3.8, 4.4 and 6.3 cm, respectively. If we assume that all alpha-particle ranges are inside the chamber, then ion production rate, cm^{-3}·s^{-1}, by alpha particles in unit time in unit volume of air will be

$$q_{a\alpha}(t) = \frac{\sum_i (I_{\alpha i} \cdot N_{\alpha i}(t))}{w_\alpha} \quad (6)$$

where: $I_{\alpha i}$ – alpha-radiation intensity per one decay of i-radionuclide (MeV decay^{-1}); $N_{\alpha i}(t)$ – alpha decay of i-radionuclide quantity in 1 s in 1 m^3 in the moment

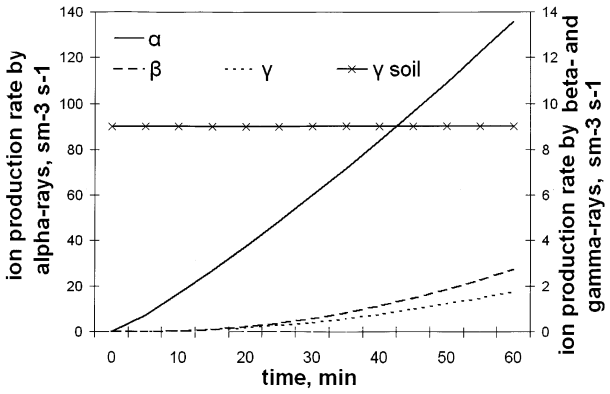

Fig. 4. Dynamics of ion production rate by different type radiation of air and soil radionuclides.

of time t, calculated by formula (5) (decays·s^{-1}·cm^{-3}); w_α – average energy for one ion pair production by alpha-particle in the air, w_α = 35 eV (3); values of $I_{\alpha i}$ were taken from [12].

Ion production rate inside the accumulative chamber due to beta and gamma radiation of ^{214}Pb and ^{214}Bi can be roughly evaluated by formula (6), evaluations are represented in Fig. 4. The average energy for one ion pair production by beta radiation in the air is 35 eV [3], gamma radiation – 33.85 eV [18]. However, such evaluations are too overrated, because only minor part of the beta particle and gamma-ray ranges formed inside the chamber will be kept within the chamber air and, consequently, quantity of interactions causing ion pair production will be small. For correct estimates, the Monte Carlo (MC) method is required.

Overall ion production rate inside the accumulative chamber (q_{in}) is caused by radiation of radionuclides in the chamber air (q_{aj}), by radiation of radionuclides contained in the soil where the chamber is installed (q_{sj}) and by cosmic radiation (q_c)

$$q_{in} = \sum_j (q_{aj} + q_{sj}) + q_c \qquad (7)$$

where j is the ionizing radiation type.

Activity of gamma and beta radionuclides ^{214}Pb and ^{214}Bi in the air of a chamber is by several digits lower, than the activity of gamma and beta radionuclides contained in the soil (^{40}K, ^{137}Cs, series ^{238}U and ^{232}Th in radioactive equilibrium with their decay products). Therefore, soil radionuclides can make a major contribution to overall ion production rate inside the chamber. Calculations of gamma-ray and electron fluxes from the soil surface and ionization produced by them are possible to be performed by the MC method for the specific soil type and conditions of experiment. In practice, a difficulty arises in input data specification. Thus, the following data are required: specific activity of all radionuclides contained in the soil, soil density, etc. The easier method to evaluate soil radionuclide contribution to value q_{in} is dose rate measurement at the experimental site and then the translation of the measured value into the ion production rate via known energy equivalents by the following relation [18]

$$0.1\ \mu\text{Sv/h} \approx 6.6\ \text{ion-pairs/cm}^3\cdot\text{s} \qquad (8)$$

Thus, soil radionuclides contribution $q_{s\gamma}$ will be in the range of 5–10 ion-pairs·cm^{-3}·s^{-1} and higher, depending on the soil type, and it will be constant for a given place of measurement. At our experimental site [28], the measured dose rate near the soil surface is 0.14 μSv·h^{-1}, that matches 9 ion-pairs cm^{-3}·s^{-1}.

Figure 4 represents the dynamics of ion production rate inside the accumulative chamber caused by ionizing radiation of radon and its decay products and by gamma radiation of soil radionuclides. As follows, the contribution of beta and gamma radiation to overall ion production rate inside the chamber does not exceed 5 ion-pairs cm^{-3}·s^{-1} after 1 h of accumulation. After 10–15 min of radon accumulation, the ion production rate by alpha radiation will exceed the background value caused by soil radionuclides. This shows the possibility of using this parameter for the measurement of radon flux density.

Consideration of thoron and its decay products contribution

The other isotope of radon – thoron (^{220}Rn) and its daughter decay products can influence the results of measurements if counting regime is used. According to works [11, 22], thoron flux density from the ground surface is by 2–3 orders greater than radon flux density value in units of activity (Bq·m^{-2}·s^{-1}).

To perform detailed analysis of potential effect, let us consider the regularities of thoron and its decay products accumulation inside the accumulative chamber and plot analogous dependences. The same Eq. (1) is used for calculations of dynamics of both radon and thoron radioactivities, where index (1) fits ^{220}Rn, and further ones fit the scheme of thoron decay, respectively. The intensity of thoron entry into the chamber is determined by thoron flux density from the ground surface. Thoron flux density 1 Bq·m^{-2}·s^{-1} is assumed for calculations.

The calculations have shown great differences in accumulation curves for ^{222}Rn and ^{220}Rn caused by various half-life periods of both parent and daughter radionuclides. For thoron and ^{216}Po, the accumulative curves are similar and become saturated in 6–7 min after chamber closing, and they remain practically constant during the accumulation period. As the half-life period of ^{212}Pb is long, its activity after 1 h of accumulation is low and it accounts 6% of thoron activity.

Figure 5 shows the calculation of dynamics of ionizing radiation yield performed by formula (3). Dynamics of alpha-particles yield is caused by nuclei accumulation of ^{220}Rn, ^{216}Po, ^{212}Bi and ^{212}Po inside the chamber. Dynamics of beta- and gamma-rays yield is caused by nuclei accumulation of ^{212}Pb, ^{212}Bi and ^{208}Tl.

The calculations of the rate of ion production by thoron and its alpha radiation decay products radiation were performed by formula (6). According to the calculations, the contribution of thoron to overall ion production rate inside the accumulative chamber is significant, it accounts for 95% during the first 5 minutes of accumulation. Therefore, while using ionizing chambers or electret ion chambers, operating in counting regime, additional technical devices or ways for thoron and its decay products separation are required (as in [17, 24]).

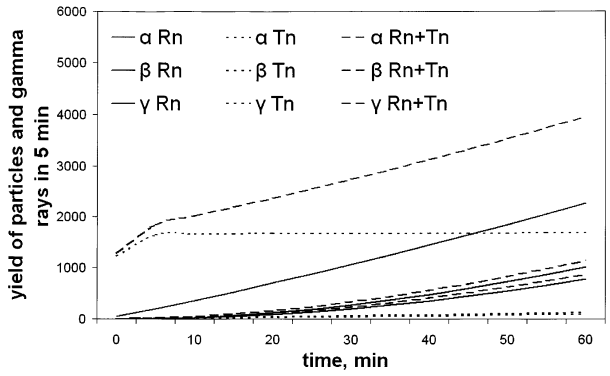

Fig. 5. Dynamics of yields of alpha-particles (upper), beta--particles (middle) and gamma-rays (lower) inside the accumulative chamber.

Conclusion

The obtained dependences (Figs. 1–5) have qualitative character, however, they are effective for quantitative predictive estimates for each particular case, as in calculations the tabulated data is used. These data are constant for a particular radionuclide, except one value – intensity of radon or thoron entry into the accumulative chamber determined by radon flux density value and chamber sizes. One can follow the obtained regularities to search and design new methods of radon flux density measurements. If radon detection is performed by alpha-radiation, the contribution of thoron and its decay products to total detector signal can reach 80%, whereas by beta- and gamma-radiation – 25% during the first 30 min of counting. The contribution of thoron to the total ion production rate is about 95% during the first 5 min of accumulation. This should be taken into account in developing new methods of measurements of radon flux density.

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