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CALIBRATION SYSTEM FOR MEASURING THE RADON FLUX DENSITY

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The measurement of radon flux from soil surface is the useful tool for the assessment of radon-prone areas and monitoring of radon releases from uranium mining and milling residues. The accumulation chambers with hollow headspace and chambers with activated charcoal are the most used devices for these purposes. Systematic errors of the measurements strongly depend on the geometry of the chamber and diffusion coefficient of the radon in soil. The calibration system for the attestation of devices for radon flux measurements was constructed. The calibration measurements of accumulation chambers and chambers with activated charcoal were conducted. The good agreement between the results of 2D modelling of radon flux and measurements results was observed. It was demonstrated that reliable measurements of radon flux can be obtained by chambers with activated charcoal (equivalent volume ~75 l) or by accumulation chambers with hollow headspace of ~7–10 l and volume/surface ratio (height) of >15 cm.

INTRODUCTION

The measurement of radon flux density from soil surface is a widely used tool for the assessment of radon-prone areas and for controlling uranium mine and mill tailings. In Russia, measurements of radon flux density on the place of future construction sites are required prior to the construction of buildings. In accordance with the standards, such measurements must have metrological support. However, there is no state standard of radon flux density in Russia.

The basic approaches for the passive measurements of radon exhalation flux density are accumulation and adsorption. The accumulation method involves the placement of a chamber called accumulator, closed at one end and open at the other, on the surface of the soil. The radon exhaled from the surface under the chamber and enters the headspace resulting in a gradual build-up of the radon concentration[1].

The basic aspects of the accumulator problem are the processes at the rim of the chamber. There are sharp variations in the concentration profiles under the chamber due to horizontal diffusion[2]. As a result, due to boundary effect phenomena, the estimated radon flux value is smaller than the real one from undisturbed soil surface[3].

Measurements of radon flux density by activated charcoal canister can be considered as a specific case of accumulation chamber. The equivalent volume of such chamber can be expressed as $V_{eq} = m{\cdot}k$, where m is the mass of activated charcoal (g) and k is absorption coefficient (l g^{-1}). Charcoal canisters are simple, passive and low cost and allow a relatively large number of measurements to be made over a reasonable time period. Their capacity to adsorb radon is reduced by adsorption of water, so they are less suitable for wet or humid environments. Following exposure, the canisters are again sealed and the activities of the radon progeny ^{214}Pb and ^{214}Bi in equilibrium with ^{222}Rn measured[1].

The main aim of this work was the creation of calibration system of radon flux density, the study of its characteristics and error estimation of calibration process.

MATERIALS AND METHODS

The design of the calibration system for the attestation of the devices for radon flux measurements is presented in Figure 1. It was constructed on the base of 200-l drum filled with quartz sand with ^{226}Ra-specific activity of <2.5 Bq kg^{-1} and calibrated ^{226}Ra source (^{222}Rn emanation rate, 0.28 Bq s^{-1}) placed in the bottom space of the system. The porosity of the sand corresponds to the effective diffusion coefficient of the radon 6.2×10^{-6} m^2 s^{-1}. The radon flux density was calculated from the measurements of total radon debit (Figure 1). To measure this value, the drum was closed by hermetic cover and air was pumped through the space above sand surface at flow rate in the range from 0.45 to 1.0 l min^{-1}. The equilibrium radon concentration was measured by radon monitor AlphaGUARD. The observed value of radon flux from the surface of 980 ± 50 mBq m^{-2} s^{-1} was in good agreement with calculated value of 960 mBq m^{-2} s^{-1} based on emanation rate of the source, dimensions of the system and filler porosity. All equations describing the process of radon concentration growth in accumulation chamber are given in the work of Sahoo and Mayya[4].

The radon concentration in the soil can be presented by the following equation:

$$\frac{\partial C_s}{\partial t} = D_{eff}\left[\frac{1}{r}\frac{\partial}{\partial r}\left\{r\frac{\partial C_s}{\partial r}\right\} + \frac{\partial^2 C_s}{\partial^2 z}\right] - \lambda C_s + \dot{A}(z),$$
$$0 \le r \le R; -H \le z \le 0, \quad (1)$$

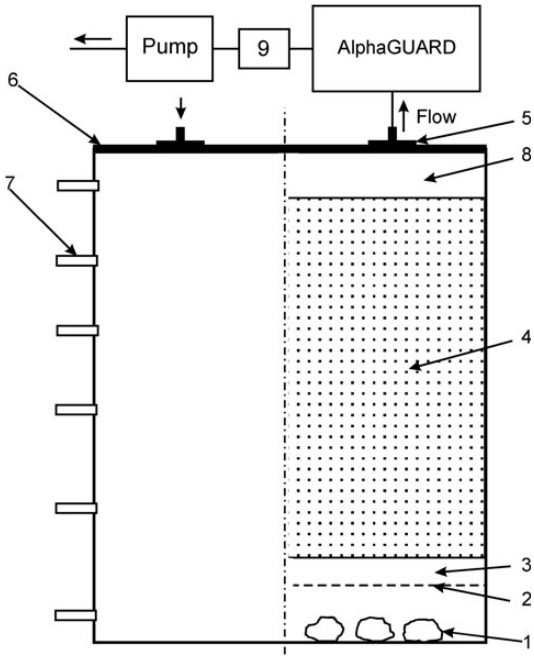

Figure 1. The design of the calibration system for radon flux measurements and the scheme of the total radon debit measurement. 1, emanation source of ^{226}Ra; 2, a metal net; 3, protective fabric; 4, quartz sand; 5 and 7, air connectors; 6, removable cover; 8, empty space; 9, rotameter.

where C_s represents radon concentration in the soil, r denotes radial coordinate, z denotes axial coordinate, D_{eff} represents effective diffusion coefficient of the radon in the soil, λ represents decay constant of the radon, $A(z)$ is source term describing the radon production in the soil, R is radius of calibration system and H represents height of space, filled by sand. The radon flux from the surface can be described by Fick's law:

$$f = -D_{eff}\left.\frac{\partial C_s}{\partial z}\right|_{z=0}. \quad (2)$$

The boundary conditions for describing radon accumulation in the chamber are as follows:

$$\begin{aligned} C_s(r, z=0, t) &= C_c(r, z=0, t) \quad \text{for } 0 \le r \le a, \\ C_s(r, z=0, t) &= 0 \quad \text{for } a \le r \le R, \end{aligned} \quad (3)$$

where C_c is radon concentration in accumulation chamber and a represents radius of accumulation chamber. The homogeneous radon concentration in the volume of accumulation chamber was assumed.

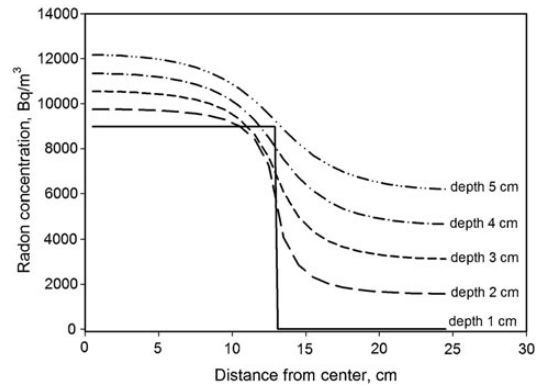

Figure 2. The spatial distribution of radon concentration on different depths under accumulation chamber.

Equation (1) considers both horizontal and vertical diffusion under accumulator. A special programme was developed to get numerically solution of this equation. As a result, it was possible to obtain radon concentration and radon flux density in any point of calibration system for selected times. Two types of initial conditions before the placing of accumulation chamber were considered:

- steady-state condition for non-emanating porous media and the constant radon source placed under the filler;
- steady-state condition with homogeneously distributed source in emanation media with a constant emanation rate.

First situations correspond to strong emanating source covered by non-active layer (radium-containing minerals covered by soil or covered uranium mine and mill tailings). Second situation characterises the soils with more or less homogeneously distributed radium concentration. The spatial distribution of radon concentration in non-emanating porous media on different depths under accumulation chamber is presented in Figure 2. As can be seen, in surface of 1-cm layer near the boundary, the radon concentration (which corresponds to the radon concentration in accumulation chamber) is higher than that in the layers of 2–4 cm. As a result, the radon leakage from the accumulation chamber to subsurface soil levels can take place near the rim of accumulation chamber. Also, the high radon concentration under the accumulation chamber decreasing the diffusion flow from soil surface to chamber volume.

The build-up of radon for different types of accumulators and parameters or soil was estimated. Also several kinds of experimental measurements were conducted as described later.

The measurement of radon flux density by an accumulation chamber with diameter of 13 cm and height

of 13 cm was performed. The accumulation chamber was placed in the centre of calibration system and was connected with pump and radon monitor on circulation scheme by plastic tubes (Figure 3).

The radon concentration growth was approximated by the following equation:

$$C_c(t) = C_c^{max}(1 - e^{-\lambda_{eff} t}), \quad (4)$$

where C_c^{max} is steady-state radon concentration in the camber under exposure time $t \to \infty$ and λ_{eff} represents growth constant of the radon in the chamber, dependent on the geometry of the chamber and diffusion coefficient of the radon in the soil D_{eff}. The parameters C_c^{max} and λ_{eff} were acquired as fitting parameters of this accumulation curve. Average radon flux density during exposure time was calculated as follows:

$$\bar{f} = \frac{C_c^{max} V}{S\lambda_{eff}}, \quad (5)$$

where V and S are volume and surface of accumulation chamber, respectively. Measurements of radon flux by charcoal canisters (6.5 cm in diameter), each containing 5 g of activated charcoal, were carried out. The equivalent volume (V_{eq}) of the charcoal canister is 75 l (equivalent height, $h = 610$ cm). Radon flux was calculated on the base of radon activity absorbed in activated charcoal A_{Rn}, time of exposure t_e and delay time t_d between the finishing of exposure and the beginning of activity counting[1].

$$\bar{f} = \frac{A_{Rn}\lambda e^{\lambda t_d}}{S(1 - e^{-\lambda t_e})}. \quad (6)$$

This equation does not take into account a saturation of radon activity in charcoal or boundary effect due to high equivalent volume.

For the assessment of spatial distribution of radon flux in the calibration system, simultaneous measurements of radon flux density from the surface by accumulation chamber and charcoal canisters were performed. Accumulation chamber was placed in the centre of emanation surface. The charcoal canisters were located around accumulation chamber at different distances (Figure 3).

RESULTS AND DISCUSSION

The numerical 2D simulating of radon accumulation in the chamber used in laboratory experiments with $a = 13$ cm and height $h = 13$ cm was conducted for two different types of sources in the range of diffusion coefficient from 1.2×10^{-8} to 6.2×10^{-6} m^2 s^{-1}. The spatial distribution of radon flux density under and outside the accumulation chamber is presented in Figure 4.

There is strong boundary effect near the rim of accumulation chamber. This effect results in the underestimating of radon flux density measured by such chamber (Table 1). Due to the influence of accumulation time on the intensity of boundary effect flow, the systematic errors also depend on the accumulation time. It was found that type of radon source distribution does not affect the systematic error. Therefore, the radon flux density measurements can be performed by accumulation chambers at any type of

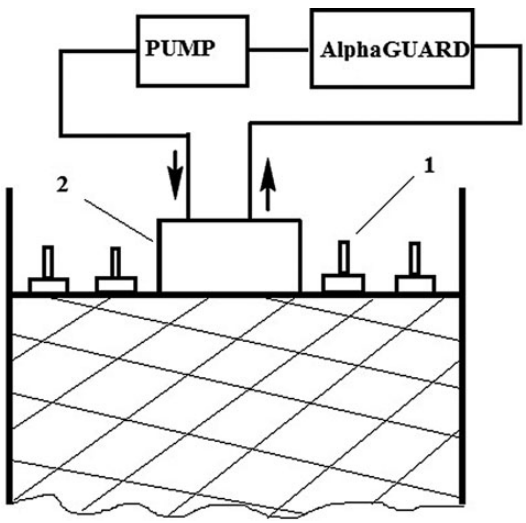

Figure 3. The simultaneous measurements of radon flux by accumulation chamber (2) and charcoal canisters (1).

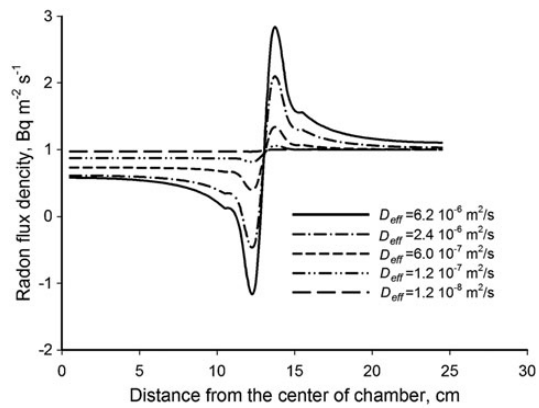

Figure 4. The spatial equilibrium distribution of radon flux density under and outside the accumulation chamber (undisturbed Rn flux 0.98 Bq m^{-2} s^{-1}).

Table 1. Dependence of the ratio of measured value of radon flux to undisturbed Rn flux density on effective diffusion coefficient of the radon in the soil (a = 13 cm, h = 13 cm) and accumulation time.

Height of accumulation chamber, cm	Effective diffusion coefficient, $m^2\ s^{-1}$		
	2.4×10^{-6}	6.0×10^{-7}	1.2×10^{-7}
1	0.29	0.39	0.57
5	0.68	0.74	0.87
13	0.84	0.89	0.95
94	0.99	0.99	1.0

Table 2. Assessment of the ratio of measured value to undisturbed Rn flux in dependence on chamber height (a = 10 cm).

Effective diffusion coefficient, $m^2\ s^{-1}$	Accumulation time, hours		
	1	2	6
6.2×10^{-6}	0.81	0.76	0.71
2.4×10^{-6}	0.88	0.83	0.78
6.0×10^{-7}	0.94	0.91	0.87
1.2×10^{-7}	0.98	0.97	0.94
1.2×10^{-8}	1.000	0.998	0.991

radium distribution in the soil. Additionally, the dependence of systematic error on the dimension of accumulator was estimated. Modelling was conducted for chamber with fixed a = 10 cm and height (h) 1, 5, 13 and 94 cm and different diffusion coefficients (Table 2).

It can be seen that, for considerable height of accumulator, there is practically no systematic error. Therefore, the measurements by charcoal canisters with a diameter of 6.5 cm and equivalent volume of 75 l were assumed to be free from systematic error due to boundary effect.

The theoretical estimations based on numerical 2-D modelling were confirmed by direct experiments on the calibration system. On the first stage, the radon flux from the emanation surface was measured by 12 charcoal canisters. The average value was 920 ± 100 mBq $cm^{-2}\ s^{-1}$ (95 % confidence interval). It is in a good agreement with calculated 960 and measured 980 mBq $cm^{-2}\ s^{-1}$ values obtained previously. These measurements have confirmed hypothesis about the absence of boundary effect influence on the results obtained by charcoal canisters.

On the next step, the radon flux density was measured by accumulation chamber with a = 13 cm and h = 13 cm. The average value was 700 ± 80 mBq $m^{-2}\ s^{-1}$ (95 % confidence interval). According to a date in Table 1, the expected value of radon flux density for such conditions should be $980 \times 0.71 = 696$ mBq $m^{-2} \times s^{-1}$.

The experiment was performed to check the influence of boundary effect on radon flux density outside the chamber. The simultaneous measurements of radon flux were conducted by accumulation chamber and charcoal canisters. The radon flux measured by chamber was 640 ± 50 mBq $m^{-2}\ s^{-1}$. The charcoal canisters placed near the chamber (4 cm from wall of the chamber to the centre of canister) showed 1130 ± 90 mBq $m^{-2}\ s^{-1}$ whereas charcoal canisters placed on a distance of 11 cm gave 890 ± 40 mBq $m^{-2}\ s^{-1}$. This experiment clearly demonstrates the boundary effect and good agreement with 2D simulating of radon flux density (Figure 4).

The system presented in the paper meets all the requirements of the primary standards: controlling of radon flux density by several independent methods that not using the standard of the same nature. Uncertainty of the radon flux density is 10 % currently, and this value can be decreased in future. The effect of soil properties such as effective diffusion coefficient affects the assessment of radon flux density, but for accumulators with hollow headspace of ~7–10 l and volume/surface ratio (height) of >15 cm, measurement error is acceptable and can be taken into consideration at analysis of calibration measurements.

CONCLUSIONS

The calibration system for metrological attestation of the devices for the measurements of radon flux density from the soil surface with the accuracy not the less than 10 % is created. The systematic errors of the measurements of radon flux density were estimated by numerical simulating of 2D diffusion processes in calibration system and accumulation chamber.

The parameters most influencing on the systematic errors are effective diffusion coefficient of the radon in the soil and height (or equivalent height) of accumulation chamber. Nevertheless, for typical effective diffusion coefficient range from 10^{-6} to 10^{-7} $m^2\ s^{-1}$, the systematic error of the measurements, conducted by the chambers with the height of >15 cm, is not exceed 15 %, which can be considered as acceptable value. The measurements of radon flux density by charcoal canisters are free from systematic errors due to boundary effect. Short-term measurements by accumulation chamber have less systematic error, but such measurements cannot be recommended because of the inevitable increase in the random error.

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