Methods of radiation and environmental studies of build-up areas and their analysis

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Abstract. The most objective assessment of radon hazard of build-up area can be the amount of radon released from the underlying rocks. In the Russian Federation, the criterion of radon hazard is the radon flux density, measured by the method of accumulation chambers. The paper presents the results of measurements obtained by the method of accumulation chamber using two measuring complexes and the method of "two depths". The measurements were performed on a testing site of Tomsk (Russia); the type of soil is loam. Studies have shown that the considered assessment methods of radon risk have significant drawbacks, because they do not allow reliable determination of the amount of radon coming from the soil surface.

1. Introduction

It was found that one of the most significant sources of radiation is radon and its decay daughter products that create more than half of the dose from all natural sources. In this regard, in the production of engineering surveys of build-up sites, the radiation and environmental studies are conducted, part of which is the assessment of radon hazard of area. However, to date there is no single approach to the assessment of radon hazard of areas. Abroad, the value of "geogenic radon potential" is widely used as a criterion for radon hazard. To determine it, they use different sets of input variables, such as: radium concentration, radon pore activity, gas permeability of soils, features of geological structure of underlying rocks [1,2]. In Russia, the value of radon flux density (RFD) is used to assess radon hazard of areas. The territory is considered safe if *RFD* does not exceed 80 mBkm⁻²s⁻¹. Despite the use of RFD as a criterion of radon hazard, the question of possible influence of measurement methods and procedures on RFD values and value of its variability has not been studied yet. The way of measuring RFD, certificated in the Russian Federation, is method of accumulation chamber. In our study, the RFD measurements were carried out by the method of accumulation chamber using measuring complexes "Alfarad+" and "Camera-01", as well as using the method of "two depths" (TD). TD method was developed in Tomsk Polytechnic University of the Russian Federation [3]. This method is based on the simultaneous measurement of volumetric activity of radon in soil pore space (hereinafter pore activity) at two depths differing twice, and diffusion model of radon transport through soil [4].

2. Measurement methods

In this study, when implementing the accumulation chamber method, measuring systems Alfarad+ and Camera-01 were used. When using complex Alfarad+ (hereinafter AC method) a chamber is installed on the surface of the soil. From this chamber the radon, accumulated during the time of exposure, is transferred to the measuring device. When measuring using complex Camera-01, radon accumulation occurs on activated carbon, followed by measuring its activity according to γ - or β -radiation of short-



lived daughter products of radon decay – the so-called method of carbon adsorbers (CA). It should be noted that the method of carbon adsorber can be considered as a modification of the accumulation chamber method, as activated carbon in the adsorber acts as an equivalent volume, in which radon accumulation also occurs.

Practice shows that the accumulation chamber method, used in the Russian Federation, has significant drawbacks. Firstly, the value of RFD measured using accumulation chambers depends on weather conditions, resulting in significant time variations of the measurement results, which can reach 100% or more. Secondly, the accumulation of radon in the chamber or on the adsorber must occur in accordance with the linear law, therefore the exposure time does not usually exceed a few hours. This condition does not allow significant smoothing out of random variations of RFD. AC method has other drawbacks - when processing the results, the exchange processes of diffusion between the air in accumulation chamber and the soil are not taken into account and radon leakage out of the chamber due to insufficient sealing of chamber joint with the soil surface. In the method of CA the impact of atmospheric humidity on the measurement results is not taken into account. This impact could be significant, as carbon actively absorbs water, and radon in its turn dissolves readily in water.

In the study we also used the method of "two depths", wherein the determining of RFD is based on the measurements of pore activity at two small depths, differing twice. In this case, according to the diffusion model of radon transport through uniform soils, the radon flux density can be calculated by formula [3]:

$$RFD = \frac{\lambda \cdot h_1 \cdot A_1 \cdot \eta}{\left(2 - \frac{A_2}{A_1}\right) \cdot \left| \ln\left(\frac{A_2}{A_1} - 1\right) \right|},\tag{1}$$

where A_1 , A_2 – pore activity of radon, measured at depths h_1 and $h_2 = 2h_1$, η - soil porosity; λ – radon decay constant. The condition of soil homogeneity, used in the obtaining of expression (1), is easy to achieve in the case of removing the upper layer of soil with thickness of 0.3...0.5 m at the points of measurement. Method of "two depths" allows obtaining a more reliable estimate of radon flux density than in the case of using accumulation chamber method when measuring RFD, as: a) methods for measuring radon pore activity provide more reliable results than methods for measuring RFD; b) radon pore activity, measured at depths of 0.4...1.2 m, is influenced by the atmosphere in a lesser degree, than radon flux density on the soil surface; c) measurements of VA is carried out in the soil with undisturbed structure, unlike measurements of RFD that are performed on the surface exposed to the strong influence of weather conditions (the surface layer is characterized by a large number of cracks occurring during drying and weathering of soils)

3. Planning of experiment

The measurements of flux density by different methods were carried out in summer 2016 on the territory located in the area of the Lagerny garden of Tomsk. In the study area, at a distance of 6 m from each other, we chose two sites with dimensions of 2m*2m (Figure 2). To provide the uniformity of soil in depth, at each site we removed the upper layer of soil with thickness of ~ 0.5 m. For measurement by TD method, two bore holes with depths of 0.4 and 0.8 m were drilled using a hand drill at each point at a distance of approximately 0.5 m. When measuring radon flux density using measuring complexes Alfarad and Camera–01, the accumulation chambers were installed in the gaps between bore holes (figure 1). In total, we carried out 95 measurements of radon flux density by the method of accumulation chamber, 60 measurements of RFD using activated carbon, 50 measurements of radon flux density using the method of "two depths" and 100 measurements of radon pore activity. Along with the preparation of bore holes, we selected the soil samples using the method of "cutting ring" to determine its porosity.

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Figure 1. Scheme of measurement points arrangement

4. Measurement techniques

"Cutting ring" method is used to determine the porosity of the soil. The soil samples were selected using a hand drill at depths of 0.1...1.5 m with a step of 10 cm. A cutting ring with an internal volume of 53.2 cm³ was pressed into selected soil samples, avoiding tool bends. After filling the ring, the soil was undercut flush with the edges and pushed onto the substrate. Then we dried the soli, determined the density of completely dried soil ρ_d and calculated the porosity by the formula: $\eta = 1 - \frac{\rho_d}{\rho_s}$, where

 $\rho_s = 2.5 \text{g} \cdot \text{cm}^{-3}$ – density of mineral soil particles.

To measure the pore activity of radon, we inserted a metal pipe with a diameter of 48 mm into each bore hole, which allowed us to exclude the contribution of radon released from the upper layers of soil, and also served to strengthen the walls of the bore holes. The samplers were dipped into the bore holes on the hanger, the rubber plugs were previously removed from the fittings of samplers (figure 2).



Figure 2. Measurement scheme of radon pore activity

Thereafter, the pipes were sealed with a resilient porous material with thickness of 5 cm, coated with polyethylene film on both sides. To prevent the ingress of rainwater into bore holes, each site was covered with a polyethylene film. The samplers were in bore hole during one day that provided the alignment of radon concentrations in bore holes and samplers. Then the samplers were removed from bore holes and radon pore activity was measured using complex Alfarad+.

In accordance with the procedure of RFD measurements using the measuring complex Alfarad+, the surface on a prepared site was leveled. After installation of accumulation chamber, we carried out the air sampling into the sampler for 20 minutes using a sampling device. After that, the air sampler was connected to a chamber Alfarad+ and volumetric activity of radon was measured in the accumulation chamber (figure 3).

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Figure 3. Scheme of air samples measurement to determine RFD. measuring device (1); air sampler (2); accumulation chamber (3)

When measuring using complex Camera 01, we used the method of radon adsorption on activated carbon with an open volume of the chamber (figure 4) and followed by measuring the activity of adsorbed radon in vitro.



Figure 4. Measurement scheme of radon flux density using carbon adsorbers. carbon (2); accumulation chamber (2).

As the sampling device the accumulation chambers AC-32 were used. We spread a smooth layer of activated carbon onto the grid. Carbon was pre-warmed in the regenerator for one hour at a temperature of 140–160 degrees, and then accumulation chamber was installed on the soil surface (figure 4). In accordance with the service manual of Camera-01, the exposure time can be 1h...10h. When carrying out the measurements, it was noticed that when exposing accumulation chamber more than 2 hours, and at relatively high ambient humidity, the carbon stored in accumulation chamber significantly increased in volume. Therefore it was decided to use the minimum exposure time of ~1h. On expiration of exposure time, the sorbed carbon in vitro was spread into detection block of beta radiation DBR-13 and radon flux density was measured.

5. The results of measurements and their discussion

Table 1 shows the results of measurements of soil porosity depending on the depth. It can be seen that soil porosity varies slightly with the depth, i.e. soil is sufficiently uniform in depth. When calculating RFD according to formula (1), we used the mean value of porosity over all measurements, which was 0.44 of relative units.

Depth, r	n	0.	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0	1.1	1.2	1.3	1.4	1.5
η , (rel. unit.)	1	0.46	0.42	0.43	0.43	0.43	0.42	0.45	0.45	0.43	0.44	0.45	0.44	0.47	0.48	0.45
	2	0.43	0.45	0.50	0.46	0.43	0.40	0.41	0.40	0.43	0.45	0.46	0.41	0.45	0.45	0.42

Table 1. Porosity of soil at different depths

In total, we carried out 95 measurements of radon flux density by the method of accumulation chamber, 60 measurements of RFD using activated carbon, 50 measurements of radon flux density using the method of "two depths" and 100 measurements of radon pore activity. We have verified the hypothesis for compliance of the measurement results of RFD to normal or lognormal distribution using the criteria of Kolmogorov-Smirnov and Shapiro-Wilk. The verification showed that radon flux density measured by the methods of AC and TD obeys the lognormal distribution, and flux density measured by the method of carbon adsorber obeys the normal distribution. Figure 5 shows the histogram for the measurements results of RFD by different methods.



Figure 5. Histogram of measurement results of RFD registered by different methods: (a) carbon adsorber method, (b) "two depths" method, (c) accumulated chamber method.

According to the results of statistical processing for the lognormal distributions, there are calculations of the means, standard deviations and coefficients of variation according to the formulas:

$$\overline{RFD} = \exp\left(\overline{\ln(RFD)}\right) \cdot \exp\left(\frac{\sigma_{\ln(RFD)}^2}{2}\right);$$
(2)

$$\sigma_{\ln(RFD)}^{2} = \frac{\sum \left(\ln(RFD)_{i} - \overline{\ln(RFD)} \right)^{2}}{(n-1)};$$
(3)

$$\sigma = \sqrt{\left[\exp\left(\overline{\ln(RFD)}\right)\right]^2} \cdot \exp\left(\sigma_{\ln(RFD)}^2\right) \cdot \left[\exp\left(\sigma_{\ln(RFD)}^2\right) - 1\right]}.$$
(4)

Table 2. The basic results of measurements of pore activity and RFD, determined using the method of "two depths" (TD), accumulation chamber (AC) and carbon adsorber (CA).

Measured value	Range of values	Mean value	Standard	Coefficient of
	x_{min} x_{max}	$\overline{\mathbf{X}}$	deviations	variation V, %
A _{0.4}	0.348	21	10	48
$(kBk \cdot m^{-3})$				
A _{0.8}	0.769	29	14	49
(kBk⋅m ⁻³)				
RFD "TD"	244	10	10.8	100
$(\mathrm{mBk}\cdot\mathrm{m}^{-2}\mathrm{s}^{-1})$				
RFD "AC"	396	39	31	80
$(\mathbf{m}\mathbf{B}\mathbf{k}\cdot\mathbf{m}^{-2}\mathbf{s}^{-1})$				
RFD "CA"	282	38	19	51
$(\mathrm{mBk}\cdot\mathrm{m}^{-2}\mathrm{s}^{-1})$				

Table 2 shows the basic results of measurement: range of values, mean values, standard deviation and coefficients of variation $V = \frac{\sigma}{r} \cdot 100\%$, where σ – standard deviation, \bar{x} – mean value.

From the data given in the table, we can see that the mean values of the results of RFD measurements by the methods of AC and CA are practically the same, which indicates the reliability of the means obtained. The coefficient of variation for AC is significantly greater than for CA that is most likely explained by a longer exposure when measuring by CA. Quite unexpected results have been obtained for the method of two depths – the mean value of RFD is four times less, and the scatter in measurement results is greater. During the measurement, it was observed that on the surface of samplers, extracted from bore holes, a large amount of moisture was accumulated. Therefore, in the final period of the study we carried out six measurements of air humidity in each bore hole – relative humidity was 93% on average (much greater than atmospheric humidity – 44%). It is known that radon is readily soluble in water. It can be assumed that radon released from the soil is actively absorbed by moisture, which leads to a decrease in its concentration in the air of soil pores. To obtain the final conclusions about effect of humidity on the measurement results it is necessary to carry out further research.

6. Conclusions

To obtain reliable estimates of the amount of radon coming to the soil surface, it is necessary to use sufficiently large exposure times.

The results of measurements of radon flux density by the methods of CA and "TD" are greatly influenced by air humidity, at which radon exposure is carried out.

Methods and techniques for measurement of flux density and radon pore activity require further study and improvement.

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