Comparative Analysis of the Measuring Results of the Radon Flux Density and Ra-226 Specific Activity for Different Soils Types

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Abstract. High concentration of radon in the premises leads to a risk of oncological diseases in the population living on the lower floors of buildings. In this regard, an assessment of the potential hazard of radon is made in the construction sites under the production of design and construction works. In the Russian Federation radon flux density measured on the earth's surface is used to estimate the potential radon hazard of the building site. The research of the relationship between the radon flux density values and the amount of radium in the soil from which radon is formed is topic of interest. The article demonstrates the comparative analysis of the measuring results of radon flux density and specific activity of Ra-226 obtained on the surface of the different soil types.

INTRODUCTION

The most significant source of radioactive radiation is radon and its daughters, which generate more than half of the dose from all natural sources [1]. Currently, in many countries, maps of geogenic radon potential are developed in order to determine the degree of radon danger for large areas [2-4]. Various databases are used to build such maps. For example, to simulate the map of the Lazio region (Italy) the data for radon volumetric activity in the soil air, the faults in the territory, gas permeability of soils and radium content were used [5]. Doses of gamma radiation, geology and gas permeability were used in mapping in Switzerland [6]. In Norway [7], the use of data on the volume activity of radon in the premises and data on the uranium content in the soils began for this purpose, since radon-222 is formed in the radioactive decay chain of uranium-238. However, these studies do not discuss the correlation between the radon release from the soil surface and the radium-226 content. Nevertheless, today there is no consistent methodology for construction such maps, since so far no parameters have been identified that uniquely correlate with the amount of radon entering buildings.

In the Russian Federation, when generation regional maps of potential radon danger, data of the uranium/radium amount in soils and of tectonic faults are used. But the regional maps of potential radon hazard are not used for assessment of radon risks in the building plots. In accordance with regulatory documents of the Russian Federation, for assessment of radon risks, the measuring results of the radon flux density (RFD) on the soil surface are used. Thus, the research of the relationship between the RFD values and the amount of radium in the soil is topic of interest. The article demonstrates the comparative analysis of the measuring results of RFD and specific activity of Ra-226 obtained on the surface of the different soil types.

EXPERIMENTAL PART

Radon flux density measurements were carried out in the spring-summer period of 2018 at three testing sites in Tomsk (terrace of the Tom River in the Lagerny Garden, Russia) and seven sites of Gorny Altai (Russia), characterized by different types of soil. The selection of the measurement area in Tomsk stem from the fact that the geological outcrops of the Lagerny Garden on the right bank of the Tom River represent a key geological section included in the international directory. On the terrace, measurements sites with different types of soils were selected (Fig. 1): loess loam (site 1), porcelain clay (site 2), and argillaceous slate (site 3) (56°27'03.6"N 84°57'00.0"E).



FIGURE 1. Layout of the radon flux density measurement sites in Tomsk

Radon flux density measurements in Gorny Altai were carried out in the valley of the Maima river, the valley of the Katun river, Gorno-Altaisk, and Kyzyl-Ozek on the following types of soil: sand-and-gravel sediments (1st (52°01'06.8"N 85°54'37.6"E), 2nd (52°01'20.7"N 85°54'19.6"E), 3rd (51°59'03.8"N 85°51'58.6"E) sites), rocky limestone (4th (52°00'02.6"N 85°53'33.6"E) site), clayey limestone (5th (52°00'15.0"N 85°54'33.6"E) site), and esite-basalt porphyrite (6th(51°56'09.6"N 85°54'50.5"E) site), and quartz rock (7th(51°54'15.5"N 85°59'28."E) site). The layout of the mentioned measurement sites is shown in Fig. 2. Studies [8-11] show that soil moisture has a profound effect on RFD measurement results. Therefore, to determine soil natural moisture soil sampling was also carried out by means of the cutting ring method at the selected sites.



FIGURE 2. Layout of the RFD measurement sites in Gorny Altai

Radon flux density measurements were performed by means of the Alfarad Plus measuring complex. In

accordance with the measurement procedure, the soil surface of the selected sites with a thickness of approximately 5 cm was removed and then the measurements sites were leveled. Then, an accumulation chamber was installed, pressing it tightly to the soil surface, and left for 15 minutes. After that, the chamber was connected to an autonomous air blower to transfer the air accumulated in the chamber to the sampler. Air sampling in the sampler was carried out for 5 minutes using a sampling device, then the sampler was connected to the measuring device (Fig. 3).



FIGURE 3. Air sampling by accumulator chamber method (1 – measuring device; 2 – air sampler; 3 – accumulation chamber)

For the determination of radium amount, soil samples were received at the sites. The mass of selected samples ranged from 0.3 kg up to 1.5 kg. The specific activity of Ra-226 depending on the sample mass was measured in the vascular geometry of "Marinelli" with a volume of 1 liter or the vascular geometry of "Denta" (truncated cone) with a volume of 250 ml using the γ -spectrometer based on hyperpure germanium semiconductor detector (CANBERRA GC2018) with a resolution of 1.85 keV and 0.85 keV for the energy of 1.33 MeV and 122 keV, respectively. Calibration was performed on the Eu-152 source with a loose density of 1 gcm⁻³. The measurements were carried out in the conditions of the radioactive balance along the most intense lines of the radon daughters Pb-214 (295.21; 351.92 keV) and Bi-214 (609.32 keV). Before measuring, the soils were thoroughly dried at a temperature of 100°C and pulverized. The holdup time of the tightly closed and sealed vessels was two to three weeks. The measurement time for one sample was one day. The statistical measurement error did not exceed 15%. The CANBERRA "Genie-2000" software was used to process hardware gamma spectrum.

RESULTS AND DISCUSSION

When conducting research in Tomsk, 64 measurements of radon flux density were obtained on the surface of three soil types: 30 measurements on the surface of loess loam, 21 ones on the surface of argillaceous slate, and 13 ones on the surface of porcelain clay.

The range of values, average values, standard deviations, coefficient of variation of the radon flux density, as well as average values of the natural soil moisture and radium content are given in Table 1. It can be seen that for loess loam the obtained average value is the typical value for this soil type, equal to 44 mBqm⁻²s⁻¹. The RFD average value for porcelain clay (59 mBqm⁻²s⁻¹) is almost two times higher than for argillaceous slate (33 mBqm⁻²s⁻¹). The relatively large variability of RFD for porcelain clay is associated with the influence of precipitation. Rainfall was regularly during the observation period. It is known that porcelain clay is moisture-retentive. Therefore, it swells after rainfall and, as a result, its gas permeability decreases. Additionally, the precipitation explains the high values of the natural moisture content, which range from 17% to 29%.

Soil type	Loess loam	Argillaceous slate	Porcelain clay
Number of measurements	30	21	13
RFD value range, mBqm ⁻² s ⁻¹	from 20 to 71	from 22 to 59	from 20 to 130
RFD mean, mBqm ⁻² s ⁻¹	44	33	59
Standard deviation, mBqm ⁻² s ⁻¹	16	10	34
Coefficient of variation, %	37	31	57
Natural moisture content, %	29	23	17
Radium content, Bqkg ⁻¹	30	160	190

The content of radium in loess loams is 30 Bqkg⁻¹, in argillaceous slate and porcelain clay is about five to six times more. However, the RFD mean for porcelain clay (59 mBqm⁻²s⁻¹) is slightly higher than for loess loams (44 mBqm⁻²s⁻¹) and even less than for argillaceous slate (33 mBqm⁻²s⁻¹).

When conducting research in Gorny Altai, 74 RFD measurements were obtained on the surface of five soil types: sand and gravel sediments, rock limestone, clay limestone, andesite-basalt porphyrite and quartzite. Table 2 presents the range, average values, standard deviations, coefficients of variation of radon flux density, natural soil moisture and radium content obtained at the sites in Gorny Altai.

Soil type	Natural moisture content, w,%	Radium content	Number of measure ments	Values range, mBqm ⁻² s ⁻¹	$\frac{\text{Mean,}}{x}$	Standard deviation, mBqm ⁻² s ⁻¹	Coefficients of variation, %
Sand-and- gravel sediment 3rd terrace	3	-	9	from 64 to 140	110	23	20
Sand-and- gravel sediment 2nd terrace	5.5	140	12	from 114 to 320	220	55	24
Sand-and- gravel sediment 1st terrace	2	-	10	from 130 to 200	170	21	12
Rocky limestone	6	180	4	from 220 to 370	292	63	22
Clayey limestone	6	180	10	from 310 to 690	500	122	24
Andesite- basalt porphyrite	5	170	17	from 500 to 1100	760	155	20
Quartzrock	3	150	14	from 540 to 1300	810	196	24

TABLE 2. Measurement results at Gorny Altai sites.

The results obtained in Gorny Altai are characterized by elevated RFD. In this regard, it should be noted that the natural moisture content of soil in Gorny Altai is significantly less than in Tomsk. The maximum RFD values on the soil surface are observed at moisture content from 4% to 7% [8, 11]. These moisture values

coincide with the natural soil moisture in the Altai Mountains. With moisture increase the radon flux density decreases significantly. An increase in soil moisture from 7.5% to 25% leads to a decrease in RFD by more than an order of magnitude: 38 mBkm⁻²s⁻¹ and 2 mBkm⁻²s⁻¹, respectively [11]. Relatively small RFD values are obtained for sand-and-gravel sediment, which are characterized by high gas permeability. These soil types have a comprehensive mineralogical and granulometric composition, therefore the average RFD values for the three terraces vary notably. The highest average values are for quartzrock (metamorphic rock) and porphyrite (magmatic rock).

The specific activity of radium for the first and third sites was not measured, since soil samples from these sites could not be pulverized due to contained in them large gravel grains. The specific activity of radium in soil samples taken at sites 2, 4, 5, 6, 7 in Gorny Altai is in a relatively small range from 140 $Bq\cdot kg^{-1}$ to 180 $Bq\cdot kg^{-1}$. However, the range of radon flux density values measured on the surface of these soils is much wider: from 110 mBqm⁻²s⁻¹ to 810 mBqm⁻²s⁻¹.

Comparing the values of radon flux density and radium content obtained in Tomsk and in Gorny Altai, it can be concluded that there is no unique dependence between specific activity of radium and amount of radon exhalation on the surface of the soil. The result can be explained by the fact that the proportion of radon entering the open pores of the soil (emanation rate) and the rate of its movement to the surface (diffusion coefficient) significantly depend on the porosity and moisture of the soil [12]. This conclusion does not recommend the use of specific activity of Ra-226 as the main criterion for the generation of regional maps of potential radon hazard. Obviously, it is necessary to take into account other typical characteristics of soils when generating regional maps of potential radon hazard, for example, open porosity and natural moisture content. It should be noted that these characteristics must be determined in the conditions of the natural occurrence of soils at the foundations of buildings. Regional maps performed in this way can be used to carry out local assessments of radon risks.

CONCLUSIONS

There is no unique dependence between the specific activity of radium in soils and the radon exhalation to its surface.

The soil type is the main factor determining the exhalation of radon from its surface.

Dependence of RFD mean on the type of soil lying at the building foundations can be used to carry out assessments of radon risks of building plots.

The regional maps generation of potential radon risks requires taking into account not only the uranium/radium content, but also other soil characteristics, for example, moisture in natural occurrence.

ACKNOWLEDGMENTS

This research was supported by Tomsk Polytechnic University CEP grant Number DPA 79/2018.

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