APPLICATION AND DEVELOPMENT OF A METHOD FOR ESTIMATING GEOGENIC RADON POTENTIAL

Robert Lakatoš[1], Sofija Forkapić[2], Ljiljana Gulan[3], Selena Samardžić[1], Dušica Spasić[3], Jelena Živković-Radovanović[3], Jan Hansman[2]

[1]Faculty of Technical Sciences, University of Novi Sad, Trg Dositeja Obradovića 6, Novi Sad, Serbia
[2]Faculty of Sciences, University of Novi Sad, Department of Physics, Trg Dositeja Obradovića 4, Novi Sad, Serbia
[3]Faculty of Sciences and Mathematics, University of Priština in Kosovska Mitrovica, Lole Ribara 29, Kosovska Mitrovica, Serbia
e-mail: laki@uns.ac.rs

Abstract
The process of radon migration from the Earth's crust is a complex process and depends on several parameters, such as: soil permeability, radium and uranium concentrations in soil, presence of cracks, variation of temperature and pressure, and other environmental parameters. Radon-priority areas are common defined in two ways, the first is by direct measurement of the radon indoor concentrations, and the other by indirect testing of radon in the soil and geological parameters, all to find a dominant transport of radon to the interior of buildings. In this study a new method for estimation geogenic radon potential was developed. Based on the percentage weight of fine fraction in soil samples and measured radon concentrations in soil, the geogenic radon potential was estimated. The proposed method was applied in two distinguished regions of Serbia with different geological substrate and the obtained results were discussed and compared in this study. Soil gas radon concentrations vary between 5 and 81 kBq/m^3 indicating variability in radium and uranium content and soil granulation related to different parent soil lithologies.

Introduction
Radon maps that aim to labels radon-prone areas in Europe are most often defined through radon in soil measurements and geological parameters [1, 2, 3]. Geogenic radon potential GRP could be estimated using the soil granulometric composition which affects soil permeability for radon and the presence of radon in the soil. The methodology of GRP determination proposed in [4] which is based on radon indexes according to the weight percentage of fine fraction in soil (particles smaller than 65 µm) and radon in soil gas concentrations (Table 1) was used. Soils with the weight percentage of the fine fraction below 15% are classified as high permeable, in the range 15-65% as medium permeable, and in the case of the fine fraction above 65% as low permeable.

Table 1. Assessment of radon index (risk) [4]

Radon index classification	Soil gas radon concentration C [kBq/m^3]		
Low	C<30	C<20	C<10
Medium	30≤C<100	20≤C<70	10≤C<30
High	C≥100	C≥70	C≥30
	Low permeability	Medium permeability	High permeability

A method that includes soil permeability estimates and radon in soil concentration measurements was used to assess radon potential for the first time in our region, mainly in Vojvodina region - the Northern Province of Serbia. Vojvodina region (Northern Province of Serbia) is a distinctly flattened lowland with altitudes of 68 to 120 m that belongs to the southern parts of the Pannonian Basin. It was mostly formed at the beginning of the Miocene by deposition of marine sediments: conglomerates, sandstones, marls, and clays. During the Pliocene, the Pannonian Sea has been transformed into the Pannonian Lake, or a system of lakes that are more like ponds and swamps. At this stage, in Vojvodina part of the Pannonian Basin deposited a large amount of sediment (paludal layers-sand, sandy siltstones, siltstones, and clay) [5]. And finally, in the Pleistocene (Quaternary era), the aeolian process during the inter-glacial formed light sediments loess - the most important parent substrate in Vojvodina, on which the agricultural land was created [6].
To examine the effect of lithological types to GRP, study was continued on three locations (Sokolica, Grabovac and Rudare) near the town Kosovska Mitrovica on the south which belongs to Vardar's zone with volcanic rocks in the geological substrate. This is a hilly terrain with an elevation from 520 to 720 m a.s.l. close located to Trepča, one of the largest industrial and mining complexes of Europe, included 40 mines, several flotation tailings, smelters and few factories. The local geology is characterized by numerous rocks like schists, sandstones, phyllites and quartzites formed in the Paleozoic era and magmatic rocks from Triassic (diabase, basalts and serpentinites). Quaternary fluvial and alluvial deposits overlap the terrain along the River Ibar valley [7]. However, vulcanite from Miocene (dacites, andesites and latites) dominate in the study area with sedimentary rocks of the Jurassic age such as marls, sandstones, shales and argillites. Hence, in the geotectonic and seismic terms examined area is characterized by different vertical movements forming a network of seismogenic faults that classified it as a moderate seismic area [8, 9].

Experimental

On 17 locations covering several geological units, the soil gas radon concentrations were accompanied by gamma spectrometry analysis of soil, indoor radon measurements (active and passive testing), and particle size distribution of soil samples. Indoor radon concentrations in selected houses were determined by several methods: passive by charcoal canisters or CR39 track detectors or active with the RAD7 (Durridge Company Inc.) radon monitor. Radon in soil concentrations were measured by RAD7 active device using sniff mode and grab protocol. The stainless soil gas probe was inserted to the depth of 80 cm from which 3.5 l of soil gas was extracted during the pumping (the airflow rate was about 0.7 l/min). Soil samples were taken from the given location, as well as stone, which was probably used in the construction of the house for gamma spectrometric analysis of radionuclide content. These samples were dried to constant mass, crushed, sieved and packed in plastic boxes of cylindrical geometry which were hermetically sealed and left for 40 days to establish a secular radioactive equilibrium. Gamma spectrometric measurements were performed with HPGe detector GMX type, with a resolution of 1.9 keV and a nominal efficiency of 32%. The passive protection around the detector is made of 12 cm thick lead. Gamma spectra were collected and analyzed using Canberra Genie 2000 software. Gamma lines of first daughter ^{234}Th were used to determine the activity concentration of uranium ^{238}U. A particle size distribution analysis was conducted using a Malvern Mastersizer 2000 Particle Size Analyzer capable of analyzing particles between 0.02 μm and 2000 μm. Based on particle size analysis the following fractions were determined: coarse sand (500-2000 μm), medium sand (250-500 μm), fine sand (62.5-250 μm), silt (3.9-62.5 μm), and clay (<3.9 μm).

Results and discussion

Based on the obtained values of the percentage weight of fine fraction in soil samples and measured radon concentrations in soil, the geogenic radon potential was estimated according to the criteria given in Table 1, and the results for the tested sites are shown in Table 2. GRP estimations are compared to maximal indoor radon concentrations measured in the neighboring houses and activity concentrations of radium, thorium and uranium detected in soil.

Table 2. Maximal indoor radon concentrations and radon in soil concentrations with radium, thorium and uranium concentrations in soil samples collected from the surrounding areas of houses with a percentage of fine fractions and GRP estimations

Location	^{222}Rn indoor [Bq/m^3]	^{226}Ra [Bq/kg]	^{232}Th [Bq/kg]	^{238}U [Bq/kg]	^{222}Rn in soil [kBq/m^3]	fine fraction (<65 µm) %	GRP
Sremska Mitrovica1	135(49)	29.8(1.4)	44(3)	< 40	32.2(2.8)	41.93	Medium
Sremska Mitrovica 2	380(6)	33(5)	47.2(2.9)	31(12)	25.1(2.5)	45.61	Medium
Sremska Mitrovica 3	133(49)	39.4(1.9)	46(10)	< 70	35(3)	45.99	Medium
Bački Petrovac	334(11)	37(6)	53(3)	25(11)	46(3)	59.49	Medium
Kulpin	144(6)	30.0(2.5)	41.3(2.1)	24(8)	12.8(1.7)	45.35	Low
Petrovaradin	173(10)	28.8(1.9)	35(6)	34(7)	10.5(1.7)	42.06	Low
Šajkaš	26(4)	33(3)	34(6)	51(13)	5.0(1.2)	51.14	Low
Beška	79(6)	24.2(1.4)	34(8)	26(21)	9.0(1.5)	40.28	Low
Novi Sad 1	110(7)	23.0(2.2)	28.8(2.6)	15(8)	21.7(2.3)	29.48	Medium
Novi Sad 2	90(6)	23.8(1.8)	24.8(2.1	< 12	5.9(1.2)	42.81	Low
Kikinda	54(6)	17.6(1.4)	27.4(2.0)	< 16	8.8(1.5)	35.27	Low
Novi Bečej	238(63)	26.3(1.7)	31.5(2.0)	37(5)	8.8(1.5)	38.44	Low
Čonoplja	262(9)	33.8(2.0)	32(10)	28(9)	12.6(1.7)	66.25	Medium
Sombor	384(78)	38(4)	45(10)	34(8)	1.1(0.6)	64.62	Low
Sokolica	362(78)	61.5(1.7)	88(3)	155(8)	81(5)	26.68	High
Grabovac	926(199)	67.9(0.9)	83(3)	134(7)	28.7(2.7)	28.78	Medium
Rudare	2843(217)	50.3(1.0)	79.4(2.8)	106(6)	7.5(1.3)	32.37	Low

The activity concentrations of natural radionuclides in three soil samples from Kosovo and Metohija are above the usual values for agricultural land in Vojvodina, for which there is a rich system of measurements [10]. Also, the particle size distributions for these three soil samples are shifted towards a larger fraction (Figures 1) and based on that higher soil permeability and higher GRP could be expected. However, on Grabovac and Rudare locations moderate and low radon concentrations in soil were detected (28.7±2.7 kBq /m^3 and 7.5±1.3 kBq/m^3, respectively). which implies the moderate and low radon risk index. Large variations in indoor radon concentration in the house in Rudare can be explained by the existence of a deep fault zone and a seismotectonic zone in the vicinity of Kosovska Mitrovica that allows the transport of radon through cracks during the mining or other seismic activities. Unlike the soil in Kosovo and Metohija, the land of Vojvodina has granulation shifted to a finer fraction and thus the assessment of the radon potential to moderate and low.

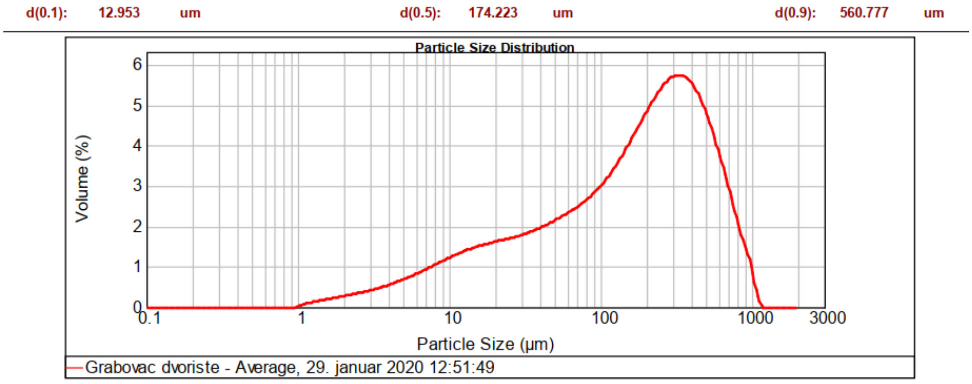

Figure 1. Particle size distribution of soil sample from Grabovac near Kosovska Mitrovica

Conclusion

The estimated geogenic radon potential of the sites shows the previously stated claims that the indoor radon concentration is not closely related to the estimation of GRP. According to obtained measurements and soil type comparison it was point out that in soils originated from the oldest (volcanic) rocks the particle size distributions are removed towards a larger fraction. unlike to youngest lithological types of soils (alluvium and loess) which particle size distributions are shifted to the fine fraction. The particle size distribution is one more factor which can be used in radon potential estimations. The need for further radon investigations in the mining area of Rudare is emphasized according to the obtained high radon levels in houses.

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