

RELATION BETWEEN INDOOR RADON CONCENTRATIONS AND GEOGENIC RADON POTENTIAL

Bistra K. Kunovska^{1*}, Todor Y. Yordanov¹, Desislava K. Djunakova¹, Dimitar I. Antonov²,
Sava N. Kolev², Kremena G. Ivanova¹, Aleksey D. Benderev²

¹National Centre of Radiobiology and Radiation Protection, 3 Georgi Sofyiiski, Str., 1606 Sofia, Bulgaria

²Geological Institute, BAS, Acad. G. Bonchev str., bl. 24, 1113 Sofia, Bulgaria

Abstract

The study presented the results of radon measurements in homes, gamma dose rate, and geogenic radon potential (GRP) in different settlements in Bulgaria. The study aims to assess the relation between measured parameters. Indoor radon measurements were carried out under the Bulgarian National Radon Plan, as follows: for the Sliven district during 2018 - 2019 and the city of Kyustendil during 2020 - 2021. The nuclear track detectors (CR-39) were used for measurements. The determination of radon concentration in soil gas, soil permeability, and gamma dose rate, were carried out under a national project by the Bulgarian National Science Fund of the Ministry of Education and Science (Contract № KII-06-H37/22/07.12.2019 г.). Measurements of radon in soil gas were performed with a soil radon monitoring system RM 2 (Radon v.o.s, Czech Republic). Soil gas permeability was performed with a RADON-JOK system manufactured by Radon v.o.s, Czech Republic, with a range of $1.7E-14$ to $5.6E-11m^2$. The geogenic radon potential (GRP) is a quantity determined by the concentration of radon in soil gas and the permeability of the soil. Direct gamma dose rate measurements were performed according to standard operating procedure with a Rados-RDS 110 portable dosimetry device. Indoor radon variations range from 23 to $1314 Bq/m^3$, and the maximum calculated GRP value is 192. The average outdoor gamma dose rate is $0.14 \mu Sv/h$, which is typical for the country. IBM SPSS Statistics, v. 23 was used for performing the statistical analysis. Statistical significance correlation was found between groups of results: radon in dwellings and GRP and gamma dose rate and GRP applying Spearman's rank correlation coefficient, which showed that GRP could be used for the prediction of indoor radon.

Keywords: geogenic radon potential, dwellings, indoor radon, gamma dose rate

1. INTRODUCTION

Radon is a major source of ionizing radiation exposure for the population and has long been known to be one of the risk factors for lung cancer [1,2]. The European Union recommends a reference level of radon in homes, workplaces, and buildings with public access. The European directive 2013/59/EURATOM defines the reference level for the average annual radon concentration in the air not higher than $300 Bq/m^3$ [3], which is included in the Bulgarian national legislation [4]. Exposure to indoor radon in dwellings constitutes a serious public health risk. Indoor radon is to a large extent dependent on geogenic radon, which is radon generated in the ground [1]. Indoor radon variation is influenced by high radon concentration and high soil permeability under buildings, as well as by their construction characteristics and the habits of the dwelling's occupants. The main source of radon concentration in dwellings is the soil beneath the building, the radon content of which varies by several kBq/m^3 to hundreds of kBq/m^3 . Besides the radon concentration in the soil, soil permeability can also vary widely.

The contribution of geogenic to indoor radon can be quantified through the geogenic radon potential. The geogenic radon potential is a quantity determined by the concentration of radon in soil gas and the permeability of the soil. A methodology for the assessment of the geogenic radon potential was prepared within the framework of a scientific National project financed by the Bulgarian National Science Fund of the Ministry of Education and Science.

Radon (^{222}Rn) is a natural gas and is formed by the radioactive decay of radium (^{226}Ra), which is a decay product of uranium (^{238}U) in soil, rocks, and water. Indoor radon relates to soil gas radon concentration, soil permeability, and radium concentration in building materials. This raises the question if it is possible to use the information for geogenic radon potential and perhaps from dose rate measurements as surrogates to determine the radon risk for areas.

With this in mind, systematic investigations concerning the radon concentration in soil gas, permeability determination, ambient dose equivalent rate measurements were performed. The results of indoor radon concentration were made under the national radon program.

The study presented the results of radon measurements in homes, gamma dose rate, and GRP in different settlements in Bulgaria and an analysis of the relation between investigated parameters.

2. MATERIALS AND METHODS

2.1. Study area

The studied area covers settlements in Sliven district and city of Kyustendil, where detailed measurements of radon concentration in dwellings were carried out. Indoor radon measurements were done under the Bulgarian National Radon Plan, as follows: for the Sliven district during 2018 - 2019 and the city of Kyustendil during 2020 - 2021.

Sliven Province belongs to two tectonic zones: the Balkan Zone (East Balkan unit) and the Srednogorie Zone. Upper Cretaceous to Mid-Eocene clayey-carbonate and clastic flysch deposits predominated in the East Balkan unit, whereas Triassic carbonates and Lower to Middle Jurassic siliciclastic and shale successions are only rarely presented [5]. Srednogorie Zone in Sliven Province is characterized by Pre-Alpine fundament (composed of Precambrian metamorphites, Paleozoic granitoids and Late Paleozoic volcanites) and Alpine successions including Triassic terrigenous and carbonate deposits and Upper Cretaceous volcanic and sedimentary rocks as well as Paleocene, Eocene, Pliocene and Quaternary siliciclastic sediments [5, 6]. Thus, Sliven Province is characterized by vast variation in rocks' age and mineral and chemical composition [7].

The article research results from 8 settlements from Sliven, presented in **Fig. 1**. The selection of settlements was carried out according to method for spatial sampling for the geogenic radon potential investigation in Sliven district, Bulgaria [7].



Fig. 1. Location of the investigated settlements in Sliven district

Geologically, the region of the city of Kyustendil is located in heterogeneous medium of igneous and metamorphic rocks covered by Quaternary sediments. The latter materials that filled the Kyustendil fault have a proluvial-deluvial character. These deposits are traced along the slopes of the surrounding mountains and are mainly represented by ridged gravels with a sandy-clay filler. These geological conditions lead to medium to low vulnerability index of Kyustendil groundwater bodies [8, 9, 10].

2.2. Indoor radon measurements

The surveys for indoor radon concentration were coordinated by the National Centre of Radiobiology and Radiation Protection (NCRRP) and carried out in collaboration with Regional Health Inspectorate Sliven and Kustendyl. Time-integrated radon concentrations were measured using CR-39-based detectors for a whole year. The experts from the NCRRP have prepared a sampling scheme based on the "The methodology for determining areas with increased radon risk on the territory of Bulgaria for the preparation of a radon map"[11]. A certain number of detectors, instruction forms, brochures, and questionnaires have been prepared for each settlement. The detectors were calibrated in the radon chamber of an accredited laboratory and the radon concentrations were evaluated under the ISO standard [12]. Analysis and processing of the results were performed in the Radon Testing Laboratory at the NCRRP.

2.3. Measurements of radon concentration in soil

The RM-2 system for measuring the radon concentration in soil gas of the company Radon v.o.s., Czech Republic was used to carry out the field measurements in the settlements. The detection principle is based on an ionization chamber operating in a current mode. The range of radon measurements in soil gas is from 3 to 900 kBq/m³, air temperature +5 to +40 °C, and humidity ≤ 70% [13]. The soil radon monitoring system RM-2 consists of an ERM-3 electrometer and a set of IK-250 sampling ionization Chambers (**Fig.2**). A self-contained operational unit, the ERM-3 electrometer, provides measurements of the ionization electric current produced by radon and its progeny in the sensitive volume of the IK-250. Radon gas was collected from a depth of 80 cm. The RM-2 system is calibrated in an internationally accredited laboratory.

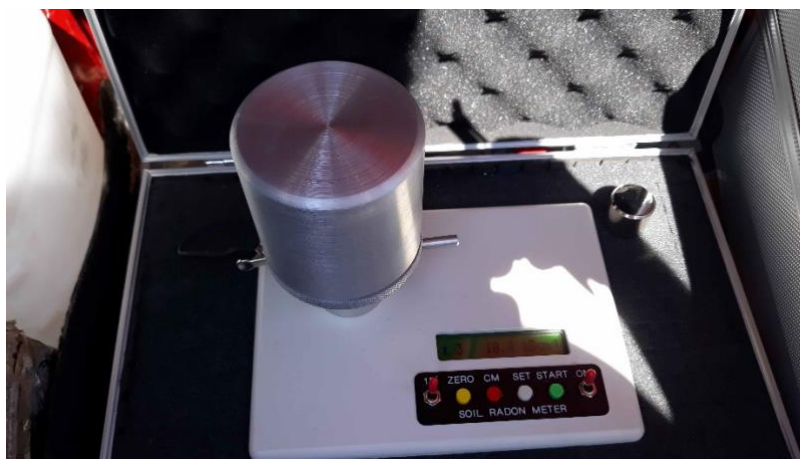


Fig. 2. ERM-3 electrometer with IK-250

Source: User manual RM-2, 2020, p.8

2.4. Measurements of soil gas permeability

Field measurements were performed with a RADON-JOK system manufactured by Radon v.o.s, Czech Republic, with a range of 1.7E-14 to 5.6E-11. [m²] with the shape factor $F = 0,149 \text{ m}$. The maximum detection limit (measured time of 2 s corresponds to $k = 5.6\text{E} -11 \text{ m}^2$) is determined by the resistance of the equipment, the minimum detection limit depends on the time spent in any given measurement

(measured time of 3600 s corresponds to $k = 1.7E^{-14} \text{ m}^2$). The principle of the RADON-JOK consists of air withdrawal using negative pressure (**Fig.3**). Air is pumped from the soil under constant pressure through a specially designed probe with a constant surface of contact between the probe head and the soil. The constant active area is created in the head of the probe by the extrusion of the tip by means of the punch wire inside the probe by an exact distance [14].



Fig. 3. RADON-JOK system

Source: Service and operation manual RADON-JOK, photo sequence, 2016, p.4

The theoretical framework for measuring soil gas permeability is based on the Darcy equation [15]. The soil is assumed to be homogeneous and isotropic and a standard state is considered. Furthermore, the air is assumed to be incompressible (pressure differences are very much smaller than the atmospheric). The airflow can be expressed by the following equation (1):

$$Q = F \times (k/\mu) \times \Delta p \quad (1)$$

where

Q is the airflow through the probe, $\text{m}^3 \cdot \text{s}^{-1}$; F is the shape factor of the probe (depending on its geometry), m; k is the gas permeability of the soil, m^2 ; μ /Pa.s/ is the dynamic viscosity of air (at 10°C $\mu = 1,75 \times 10^{-5} \text{ Pa.s}$); Δp is the pressure difference between surface and the active area of the probe, Pa [15].

2.5. Geogenic radon potential

The geogenic radon potential is a quantity determined by the concentration of radon in soil gas and the permeability of the soil. The purpose of the GRP is to quantify „what the ground supplies in terms of radon“ and is an indicator of expectation R_n in terms of indoor radon [16,17]. In the present study, for the assessment of GRP (dimensionless value) was applied a function, which could be expressed in radon concentration in the soil and soil permeability [16,17]. The used GRP equation (2) was developed semi-empirically to allow optimal prediction of indoor radon [17].

$$GRP = \frac{C_{Rn}}{-\log_{10} k - 10} \quad (2)$$

where C_{Rn} is the radon concentration in the soil, kBq/m^3 , and k is the permeability of the soil, m^2 .

2.6. Gamma dose rate measurements

Direct gamma dose rate measurements were performed according to standard operating procedure. A portable Rados-RDS 110 diametric device was used to measure the parameter. The dosimeter measuring range is 0.05 $\mu\text{Sv/h}$ to 99.99 mSv/h in the energy range of 50 keV to 1.25 MeV at air temperature -25 to +55 $^{\circ}\text{C}$. It covers the majority of significant gamma radiations emitted from terrestrial sources. The calibration accuracy is $\pm 2\%$ (2σ), according to the Calibration Certificate from 2023. A one-minute integrated count technique is a practical survey procedure for equipment. The relative uncertainty of the method was assessed to be 12%. The gamma dose rate measurements were taken in air 1 m above ground. About 10 readings were taken at each point, and the ambient mean was considered as the representative value of the gamma dose rate for the objects.

2.7. Statistical analysis

IBM SPSS Statistics, v. 23, and JASP, v.0.18.3 was used to perform statistical analysis. In order to evaluate the relationship between the studied parameters, the measurements are grouped as follows: indoor radon results, measurements of radon in soil gas and gamma dose rate, and GRP data. Descriptive analysis and Spearman's correlation coefficient are applied for data processing. Correlation is significant at the 0.01 level. The results of the Spearman's correlation coefficient calculation can be interpreted based on Cohen's scale presented on **Table 1** [18]. The results of the correlation analysis were used to describe how strong the level of relationships between the variables is.

Value (absolute)	Interpretation
$0.10 < \rho \leq 0.30$	Weak
$0.30 < \rho \leq 0.50$	Average
$0.50 < \rho \leq 0.70$	Strong
$0.70 < \rho \leq 1.00$	Very strong

Table 1. The interpretation of Spearman's correlation coefficient [18]

3. RESULTS AND DISCUSSION

The descriptive statistic of passive measurements of indoor radon concentration in dwellings (127 number), gamma dose rate with abbreviation GDR (54 number), the radon concentration in soil (54 number), permeability on soil (54 number), and assessed GRP (54 number) is presented in **Table 2**.

	Indoor radon, Bq/m^3	Radon in soil gas, kBq/m^3	GDR, $\mu\text{Sv/h}$	Permeability m^2	GRP
Arithmetic mean	223.4	25.6	0.14	1.04E-11	25.7
Standard deviation	237.6	20.0	0.03	1.01E-11	28.7
Geometric mean	179.8	20.5	0.14	5.11E-12	17.9
Median	131.5	20.6	0.14	6.81E-12	17.9
Maximum	1313.5	105.7	0.19	3.72E-11	191.7

Table 2. Descriptive statistics of measured parameters

Indoor radon variations range from 23 to 1314 Bq/m³, with average arithmetic means (AM) of 223 Bq/m³ and geometric mean (GM) - 180 Bq/m³. The average outdoor gamma dose rate in air is 0.14 µSv/h, which is typical for the country.

The average arithmetic mean of permeability on soil was 1.04E-11, according to our methodology for assessment the permeability is high. This means that if radon concentration in the soil is high, it could quickly reach the Earth's surface and accumulate in enclosed spaces such as buildings.

The calculated average value of radon concentration in soil gas was AM=26 kBq/m³ with a standard deviation of 20 and the geometric mean was 21 kBq/m³. The maximum measured result of radon concentration in soil from our survey is 105.7 kBq/m³, while the maximum radon value in soil gas found in an investigation in Estonia is 1806 kBq/m³ [19]. The geometric mean for radon in the soil of the Estonian survey was GM =31 kBq/m³ [19]. Results of similar measurements of radon concentration in soil gas were obtained in other international studies as follows: The geometric mean for radon in the soil of the Estonian survey was GM =31 kBq/m³ [19]; in a study in Northern Romania, it is found that geometric mean for radon in soil 28.4 kBq/m³ [20]; in Jordan desert in AlMafrq districts was found arithmetic mean of radon concentration in soil - 7.53kBq /m³ [21].

In our research, the median of geogenic radon potential was evaluated to be 18 and the average arithmetic means was 26. In a German study, the AM of GRP was 36 and the median was 21 [16]. The German database (status 30 November 2019) consists of 4448 locations with Rn in soil gas and soil gas permeability data [16].

A non-parametric Kolmogorov-Smirnov test was applied to examine the distribution of the data. A log-normal distribution of the results was found for parameters: indoor radon (**Fig.4a**), the radon concentration in soil gas, and GRP (**Fig.4b**).

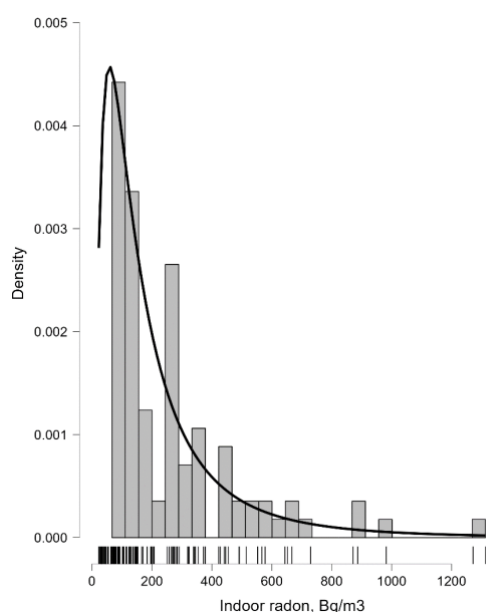


Fig.4a. Indoor radon data distribution

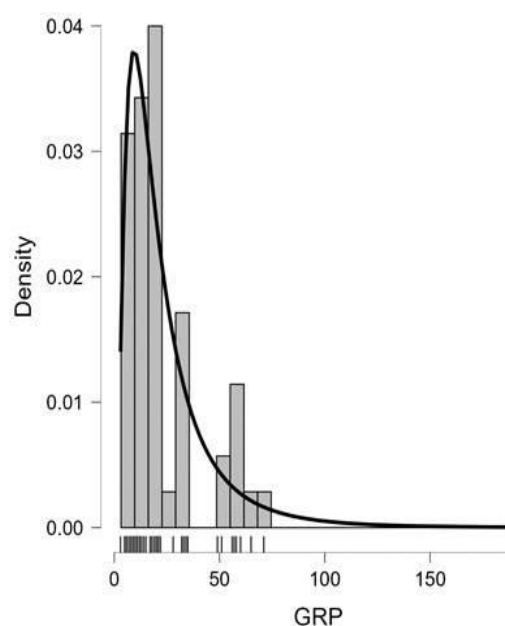


Fig.4b. Geogenic radon potential data distribution

To assess the relation between measured parameters, Spearman's rank correlation coefficient was applied (denoted by ρ). It is a nonparametric measure of rank correlation (statistical dependence between the rankings of two variables). The correlation matrix with investigation parameters and the results Spearman's coefficient is presented in the **Fig.5**.

A very strong relationship was established between the results of radon concentration in soil gas and geogenic radon potential ($\rho=0.786$) and strong for soil permeability ($\rho=0.590$). These strong relationships were expected because the GRP is directly proportional to soil gas radon concentration and inversely proportional to soil permeability.

The correlation between the gamma dose rate in the air ($\rho=0.542$) and GRP was strong since the dose rate depends on the presence of radium in the soil from which the radon is produced. On the other hand, a weak correlation of gamma radiation dose rate with radon concentration in buildings is observed ($\rho=0.209$). This result is expected because the dose rate in the building depends on the presence of radium in the building materials.

An average correlation was also established between GRP and indoor radon concentration ($\rho=0.410$). The relatively low established correlation suggests that radon in homes is affected not only by radon in the soil but also by other factors that have been investigated in numerous studies. Nevertheless, the results confirm our hypothesis of a relationship between radon in soil and radon in buildings. Therefore, making a geogenic map could help for finding the radon priority areas. On the other hand, the assessment of the radon index of the construction site makes it possible to take preventive measures for the specific building, which would not allow radon to penetrate into it.

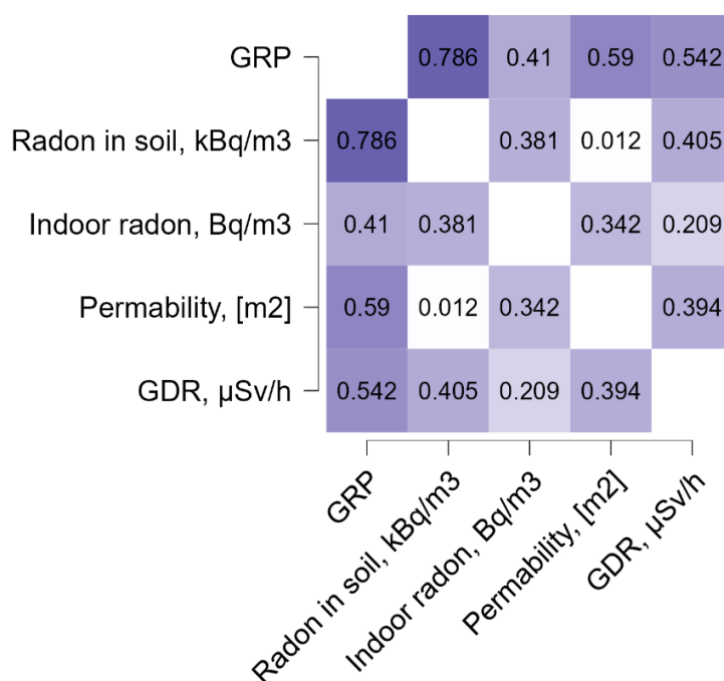


Fig. 5. Correlation matrix on Spearman's rank coefficients

The scatter plots presented in the **Fig. 6a** and **Fig. 6b** show the distribution of the relationship between the data of GRP and radon in soil gas and indoor radon respectively.

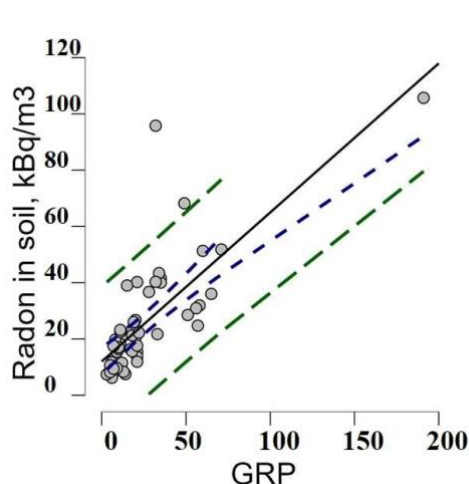


Fig. 6a. Scatter plot between the data of GRP and radon in soil gas

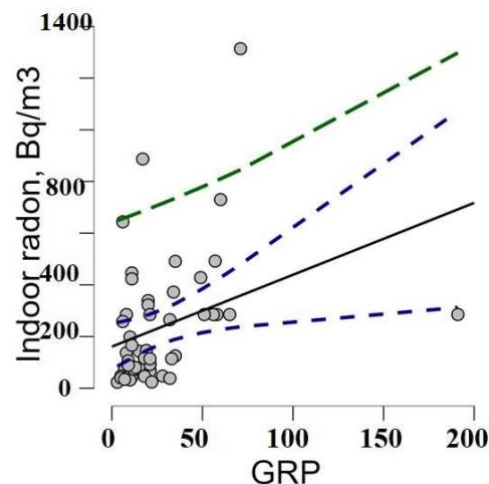


Fig. 6b. Scatter plot between the data of GRP and indoor radon.

In the figures, the confidence interval (95%) in blue and the 95% prediction interval in green are presented.

4. CONCLUSIONS

Radon gas is one of the biggest air pollutants from natural radioactive sources. Results from our study show that the average arithmetic mean on indoor radon was 223 Bq/m³ and GRP was 26.

Statistical significance correlation was found between data of GRP and: indoor radon ($\rho=0.410$), radon concentration in soil gas ($\rho=0.786$), soil permeability ($\rho=0.590$), and gamma dose rate ($\rho=0.542$) applying Spearman's rank coefficient. The correlation showed that GRP could be used for the prediction of indoor radon. This is evidence of a relationship between the amount of radon generated in the ground and the amount of radon that enters dwellings.

Additional analysis should be performed based on the rock composition of the investigated sites and an assessment of the influence of geology. These studies will serve to establish a connection between the GRP and the geogenic features on the territory of Bulgaria. This research may serve as a base for further detailed investigation of the geogenic radon potential.

ACKNOWLEDGMENTS

This study presents a part of the research conducted within the national project „Geogenic potential for evaluation of radon index in construction of new buildings“ is supported by the Bulgarian National Science Fund, in the framework of Grant No KII-06-H37/22/07.12.2019 r.

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