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ENVIRONMENTAL RADIOACTIVITY WITH RESPECT TO GEOLOGY OF SOME SERBIAN SPAS

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Abstract

This study was performed in order to investigate gamma dose rates, radon exhalation rates and soil radioactivity with respect to geology. Fifteen locations in three Serbian spas were under investigation. Gamma dose rate was measured by Radex RD1503^{+} monitor. GIS technology was applied for the interpretation of the results. HPGe detector was used for gamma-spectrometry determination of soil radioactivity. Radon exhalation rate was determined using RAD7 device. The correlation was examined between radon exhalation rate and ^{226}Ra. No correlation was found between calculated effective dose from radionuclides in soil and measured ambient gamma dose rate.

Keywords: Soil; Radon exhalation rate; Gamma dose rate; Geology; Correlation

1. Introduction

The preliminary information of radioactivity levels in an area could be given by the measurements of gamma dose rate. The ambient background gamma dose rate shows variations in time and variations from place to place regarding local feature of geology.

Ambient gamma dose originates from natural and anthropogenic radiation. Natural gamma radiation consists of cosmic radiation and the radiation of radionuclides present in the air, soil and on the ground surface [1]. Cosmic radiation consists of primary radiation (mainly protons) and secondary radiation (generated by interaction of primary radiation and atoms in the atmosphere at altitudes about 10 km above sea level). Average worldwide effective dose (at the sea level) from directly ionizing and photon component is 32 nSv h^{-1}, and 7.8 nSv h^{-1} from neutron component [2]; latitude effects on charged particles and photon components of cosmic fluxes are limited to 10% [1]. Gamma radiating cosmogenic radionuclides (^{7}Be and ^{22}Na) and radon progenies occur as natural radionuclides in air (airborne gamma radiation). Contribution from radionuclides in air to the dose rate is very small; contribution of ^{7}Be and ^{22}Na is far below 1 nSv h^{-1}, and average contribution from outdoor radon and progenies is 2.5 nSv h^{-1} [3].

Natural radiation in soil and on the ground surface originates from primordial radionuclide ^{40}K and radionuclides in the series of ^{238}U, ^{235}U and ^{232}Th, which are present in all rocks and soils. The radionuclides are often inhomogeneously distributed due to diverse composition of soil. External dose rate can be enhanced in high natural background areas, such as in Brazil, China, India, Egypt, etc. [1]. Worldwide average contribution to external dose rate from terrestrial gamma radiation is estimated to be 480 µSv y^{-1} (410 µSv y^{-1} indoors and 70 µSv y^{-1} outdoors) [2].

The anthropogenic radiation appears after nuclear weapon tests and nuclear accidents. Anthropogenic radiation mainly relates to ^{137}Cs and its deposition after Chernobyl accident. The contribution from anthropogenic radiation source could be difficult for dose estimation; a precise determination for certain geographical area is impossible, due to different patterns of fallout deposition and uneven distribution. It is mainly unknown whether the surface/profile soil is undisturbed or disturbed, as well as whether the migration of ^{137}Cs through the different soil types is slow or fast.

On the other hand, gamma spectrometry analysis can give more reliable information of specific activities of natural and anthropogenic radionuclides in soil. Some studies have established strong correlations among dose rate, geological background and soil types [4-6]. A significant geological influence on terrestrial gamma dose rate was observed by some authors [7]. The measured doses varied according to geological background and soil types [6].

There are a few aims of this research. At first, measurements of ambient dose equivalent rates in study area were performed in order to get preliminary information about background gamma radiation nearby spas' springs. Gamma spectrometry analysis was done in order to establish the soil radioactivity, to make dose estimation and radiological risk assessment. A comparison was done between the measured and calculated dose rate values. At least, radon exhalation rates were measured and the correlation with ^{226}Ra activity in soil was investigated.

2. Materials and Methods

2.1 Geology of study area

Study area is located in south part of Serbia, in the municipality of Kuršumlija, and includes three famous spas – Kuršumlijska Spa, Prolom Spa and Lukovska Spa. Since this area is characterized by diverse geology, it provides particular conditions for background gamma radioactivity research (Fig.1). Kuršumlijska Spa is located at 442 m above sea level and it has ten natural mineral water sources with temperature of 37º to 59 ºC [8]. Prolom Spa is located at 598 m above sea level; the temperature of thermal water sources ranges from 26-34°C, and the pH value is about 8.5 [8]. Lukovska Spa is located on the eastern slopes of Kopaonik mountain at an altitude of 681 m (the highest spa in Serbia) and it is the richest in thermo-mineral water sources in Serbia. The water temperature of 13 sources ranges from 20-68 °C [9]. Lukovska Spa is characterized by a subalpine climate, where the release of significant amounts of heat from thermal springs affects the microclimate of the spa.

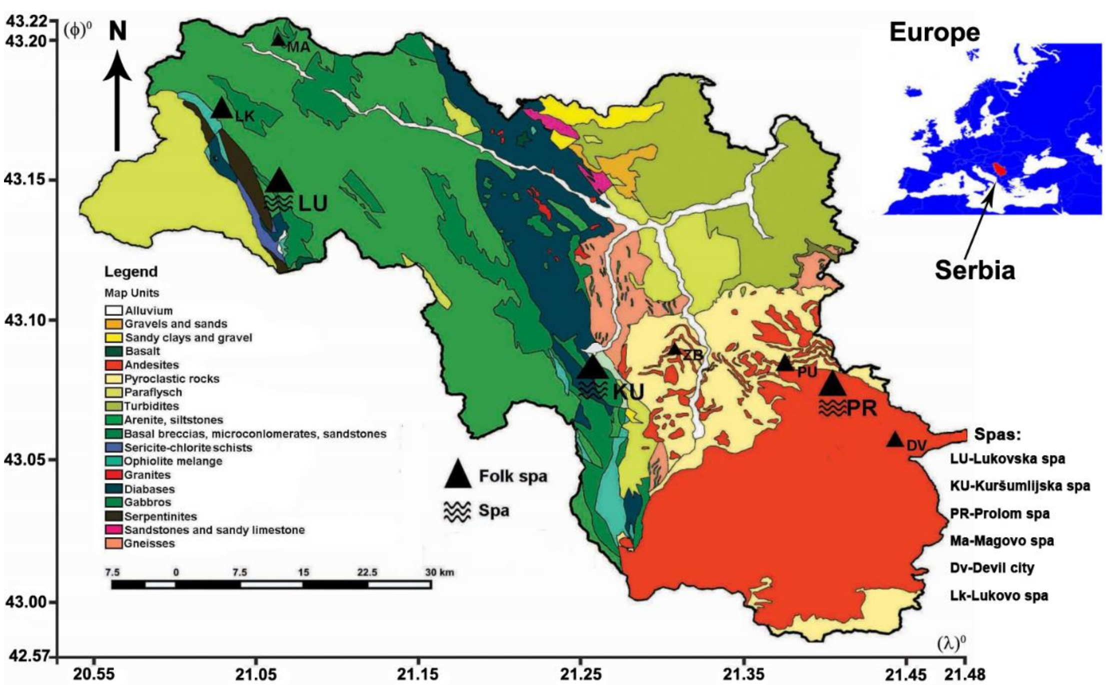

Fig. 1 Redesigned and vectorized geological map of the Municipality of Kuršumlija (Serbia)

In geology terms, a study area is a complex entity comprising at least three belts differing in the time span of existence and composition. The oldest rocks are high-grade metamorphic rocks of Proterozoic age. The most abundant are biotite and two-mica gneisses formed under the garnet-amphibolite facies. Mesozoic products include very rare Triassic, Jurassic and the most widespread Cretaceous rocks. Middle Triassic schists, sandstone and sandy limestones are distributed within a narrow and by faults interrupted zone Northwestern from Kuršumlija. Upper Jurassic ultrabasic and basic rocks with small granitoid bodies appears as discontinuous zones stretching NNW-SSE. Ultrabasic rocks are predominantly serpentinized harzburgites and serpentinites, locally hydrothermally altered and ore-bearing. The latter is particularly common in tectonized areas, but also occur as small bodies in ophiolite melange [10]. Gabbro, dolerite and diabase build larger masses in Kuršumlija ophiolitic zone or occur as small blocks in the melange. Diabase is of notable wider distribution. Their contact with adjacent rocks is always tectonic. A relatively small leucocratic granite intrusions into the Kuršumlija ophiolitic massif, derived through melting off (meta) sedimentary and immature volcanic/clastic rocks can be observed along faults [11]. Ophiolite melange consists of various sedimentary, igneous and metamorphic rocks. Part of melange, southern from Lukovska spa underwent weak metamorphism and transited into sericite-chlorite schists (Scose). Sedimentary rocks in melange include siltstone with small amounts of a sandy component in which are placed rounded fragments of silicified limestones, diabase, spilling, Upper-Jurassic limestones, greywackes and cherts. Sedimentary-volcanogenic units of the Upper Oligocene age lie transgressively over crystalline schists at borders of the Lece Massif. Conglomerates decline upward in respect to fine-grained sediments. Poly-phase volcanic activity left behind one of the largest Tertiary volcanic provinces in Serbia - the Lece Volcanic Complex, which covers an area of around 700 km^2 with characteristic calderas [12]. According to Fig. 1 and geological map [12] each spa could be characterized by diverse geological units as follows:

1) Kuršumlijska Spa – diabases, gabbros, basalt, paraflysch;

2) Prolom Spa – granites, andesites, pyroclastic rocks;

3) Lukovska Spa – basal breccias, sandstones, sericite-chlorite schists, serpentinites.

2.2 Sampling and method of measurements

2.2.1 Ambient dose equivalent rates (ADER)

In this study outdoor gamma radiation levels were measured at the height of 1m above ground by Geiger counter Radex model RD1503$^+$ supplied by QUARTA-RAD. Radex monitor operates in range from 0.05-9.99 μSv h^{-1} with measuring uncertainty of ± 15% [13]. Thirty one measurement points were randomly selected in the study area according to the vectorized geological map (Fig. 1) in the spring of 2016; four measurements were performed at one measuring point and then averaged to a single value. There was no precipitation on the day of the measurements, neither in the previous 2 days. Global Positioning System (GPS, GARMIN eTrex 10) was used to determine the geographical coordinates; altitudes of measured points ranged from 433-774 m (the error of measurement system ranged from 3 to 5 m).

Before obtaining the layout of geothermal sources, the elevation map of Serbia (Digital Elevation Model, DEM) was made; the height zone of the research area is determined. The USGS service (earthexplorer.usgs.gov) was used before downloading the satellite data for the elevation. Raster data in the form of Aster Global Dem extensions was downloaded from the above server, which were later unpacked and cropped for the territory of the municipality of Kuršumlija [14, 15]. In this research, we used advanced GIS methods for determining a distribution of gamma dose rate. In this case, we successfully adapted numerical kriging grid method for determining the dispersion of gamma dose rate. GIS (Geographical Information System) and data modelling in combination with geospatial analysis became a very powerful tool for calculating and describing some properties of radioactivity in an area. Semi-ordinary and ordinary kriging method were employed through QGIS (Quantum GIS) and SAGA (System for Automated Geoscientific Analyses) into special tools of Spatial Analyst. The priority is given to ordinary kriging, semi-kriging, global-kriging and interpolations methods since it includes autocorrelation (statistical relationship) between the measured points [16].

2.2.2 Gamma spectrometry analysis

Taking into account that each spa has different geological formations (Fig.1), fifteen soil samples were taken to the depth of 15 cm (five samples from each spa) at the same time when ambient dose equivalent rates were measured. The sampling was performed according to IAEA recommendation [17]. Prepared soil samples were hermetically sealed in 450 ml Marinelli beakers and left for more than 4 weeks to achieve secular equilibrium between ^{226}Ra and its daughters.

Table 1 Gamma lines used for measurement of radionuclide specific activities

Radionuclide	Progeny	Gamma energy [keV]	Intensity [%]
^{226}Ra	^{214}Pb	351.9	37.1
	^{214}Bi	609.3	46.1
	^{214}Bi	1764.5	15.9
^{232}Th	^{228}Ac	338.3	12
	^{228}Ac	911.1	29
	^{228}Ac	968.9	17.5
	^{208}Tl	338.3	86
	^{208}Tl	860.6	12
^{40}K		1460.7	10.7
^{137}Cs		661.6	84.6

The specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were determined using coaxial HPGe detector (GEM30-70, ORTEC). The detector has relative efficiency of 30% and energy resolution (FWHM) of 1.85 keV at 1.33 MeV (^{60}Co). Detector calibration was performed using a calibration source of a Marinelli mixture provided by the Chech Metrological Institute, Inspectorate for Ionizing Radiation (type MBSS 2 containing eleven radionuclides: ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, ^{203}Hg, ^{152}Eu). Gamma-activity of each sample was measured for 6 h. The specific activities of radionuclides were determined using gamma-ray lines presented in Table 1. The activities of ^{226}Ra and ^{232}Th were calculated as weighted average specific activities of their decay products.

2.2.3 Dose and risk assessment from radioactivity

The absorbed dose rates $\dot{D}$ (nGy h^{-1}) in the air due to natural and artificial radionuclides were estimated according the following formula [2, 18, 19]:

$$\dot{D} = 0.462 \cdot A_{Ra} + 0.604 \cdot A_{Th} + 0.0417 \cdot A_K + 0.03 \cdot A_{Cs} \quad (1)$$

where A_{Ra}, A_{Th}, A_K and A_{Cs} are specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in soil, respectively.

The calculated values of $\dot{D}$ (nGy h^{-1}) were converted to effective doses D_E (μSv y^{-1}) by multiplying with 0.7 Sv Gy^{-1} (conversion coefficient) and 1750 h (annual time for exposure outdoors).

Excess lifetime cancer risk (*ELCR*) was calculated using the D_E (μSv) and life expectancy LE (estimated to 70 y) according to following formula [20]:

$$ELCR = D_E \cdot LE \cdot RF \tag{2}$$

where RF = 5.5·10^{-2} Sv^{-1} is fatal cancer risk per Sievert for stochastic effects of radiation to the whole population [21].

2.2.4 Radon exhalation rate

Radon exhalation rate was determined by using a chamber connected through two vents to the inlet and outlet of RAD7 device (produced by Durridge, Massachusetts, USA, with absolute measurement accuracy of 5% and nominal sensitivity of 0.013 cpm/(Bq m^{-3})). A soil sample was placed in the chamber and the Rn concentration in the chamber was measured for 7 days, in cycles of 1 h. A detailed description of the measuring procedure was given by Stajic and Nikezic [22]. The growth of the Rn concentration (Sample No. 8) in the chamber is illustrated in Fig. 2. Radon exhalation rates were obtained by fitting the experimental data to the following equation [22]:

$$C(t) = \frac{E_{Rn}\, A + M + C_{\mathrm{ext}}\lambda_{\mathrm{L}} V}{(\lambda + \lambda_{\mathrm{L}})V}\left(1 - \mathrm{e}^{-(\lambda+\lambda_{\mathrm{L}})t}\right) + C_0\, \mathrm{e}^{-(\lambda+\lambda_{\mathrm{L}})t} \tag{3}$$

where $C(t)$ and C_{ext} are radon activity concentrations inside and outside the chamber, respectively; C_0 is initial radon concentration (t = 0); E_{Rn} is the Rn exhalation rate; A is the mass of a sample; V is free volume of the chamber; M is the system background caused by a possible contamination of the measuring system itself; λ and λ_{L} are the radioactive decay constant of ^{222}Rn and the leakage rate, respectively.

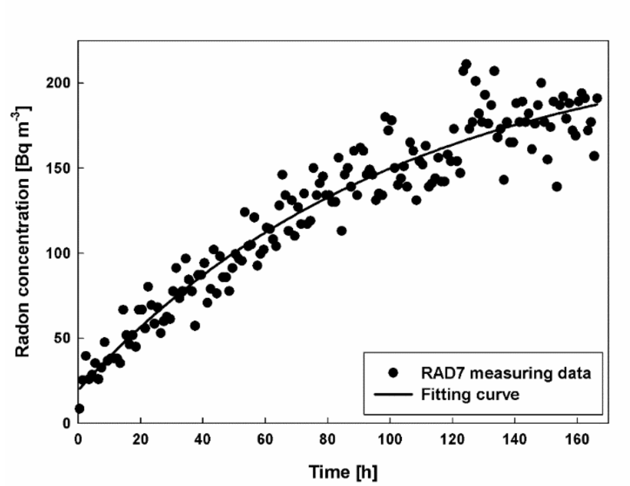

Fig. 2 The growth of radon concentration in the chamber

3. Results and discussion

Spatial distribution of gamma dose rates in the municipality of Kuršumlija is presented in Fig. 3, using GIS approach. SPSS 20.0 software was used for statistical analysis. Descriptive statistics of ambient dose equivalent rates (ADER) is presented in Table 2.

Table 2 Descriptive statistics of ADER, specific activities of radionuclides, radon exhalation rates and doses

		Radioactivity							
	ADER	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	**E_{Rn}**	$\dot{D}$	D_E	*ELCR*
	[nSv h^{-1}]	[Bq kg^{-1}]				[mBq kg^{-1} h^{-1}]	[nGy h^{-1}]	[μSv y^{-1}]	10^{-4}
Min	73	9	15	263	BDL	20	23.9	29.3	1.1
Max	170	37	53	700	38	95	73.5	90.1	3.5
Median	120	23	28	411	3.7	45	46.5	57.1	2.2
Mean	119	22	31	420	8.2	50	47.0	57.6	2.2
SD	23.7	7	11	128	10.6	23	13.2	16.2	0.6
Skewness	-0.14	0.05	0.65	0.99	1.96	0.60	0.28	0.27	0.31
Kurtosis	0.04	0.96	-0.36	0.42	3.85	-0.39	-0.24	-0.25	-0.12

BDL – below detection limit

The results ranged from 73-170 nSv h^{-1} with a mean value of 119 nSv h^{-1}; a similar value of median and low skewness indicated normal distribution. These values are comparable to the averaged ambient dose equivalent rate (from 25 monitoring sites) in Serbia which was between 81 and 142 nSv h^{-1} in 2015, according to the data from Serbian Radiation Protection and Nuclear Safety Agency [18]. There are no significant differences in gamma dose rates of measured points (Table 2), although it can be expected according to different geological features and the fact that gamma dose rates increase with altitude [2]. Almost equal spatial distribution of ambient dose equivalent rate is notable from Fig. 3, although each spa is located at different elevation; the mean values for Kuršumlijska Spa (433-454 m), Prolom Spa (548-629 m) and Lukovska Spa (677-774 m) were 110, 117, 124 nSv h^{-1}, respectively.

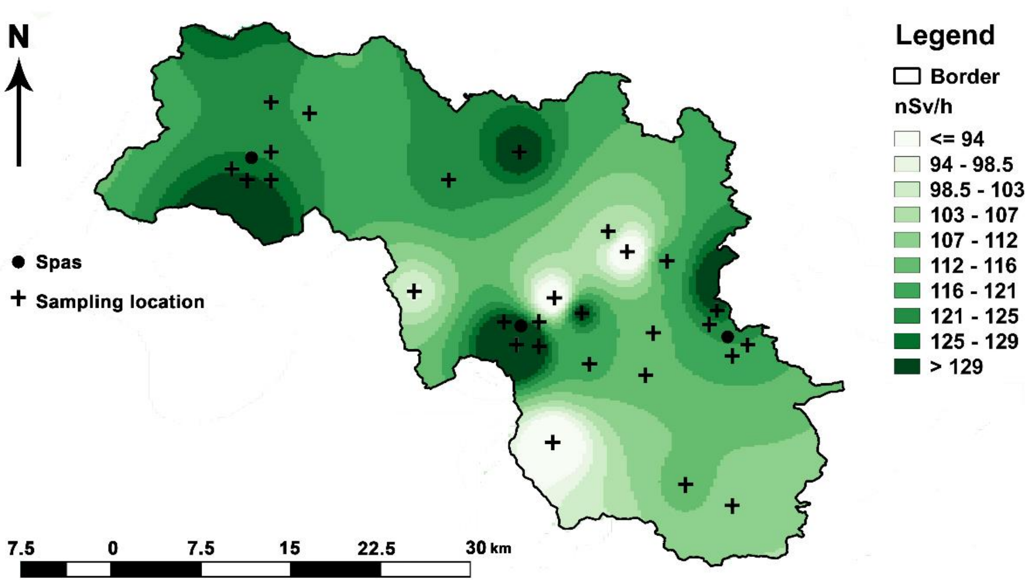

Fig. 3 Spatial distribution of gamma dose rate of Kuršumlija municipality with contour lines

The detail results of soil radioactivity are presented in Table 3. The calculated average specific activities of ^{226}Ra, ^{232}Th and ^{40}K are 22, 31 and 420 Bq kg^{-1}, respectively (Table 2). These results are comparable with the average worldwide values [2]. There is no statistically significant difference among the three spas in regard to the mean specific activities of radionuclides. All obtained values of specific activities are typical for soil samples, and there is no difference compared to values reported in other studies in Serbia [24-27]. The average value of ^{137}Cs (8.2 Bq kg^{-1}) is comparable with values of 8.7 Bq kg^{-1} recently measured in the mountain region of Stara Planina [28] and lower than the values previously measured in other regions of Serbia [29-31]. Descriptive statistics of absorbed dose rate $\dot{D}$, effective doseD_E, and excess lifetime cancer risk *ELCR* are also presented in Table 2.

Table 3 Specific activities of radionuclides and radon exhalation rates in soil

Sampling Location	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	E_{Rn}	Altitude	Latitude	Longitude
	[Bq kg^{-1}]				[mBq kg^{-1} h^{-1}]	[m]	(N)	(E)
Kuršumlijska Spa 1	12	20	347	11	37	454	43°03'333"	21°14'960"
Kuršumlijska Spa 2	22	29	469	15	45	450	43°03'401"	21°15'148"
Kuršumlijska Spa 3	25	27	473	38	75	447	43°03'413"	21°15'293"
Kuršumlijska Spa 4	23	26	445	4.1	33	454	43°03'525"	21°15'493"
Kuršumlijska Spa 5	26	39	644	2.4	36	433	43°04'001"	21°15'774"
Prolom Spa 1	20	33	416	1.3	57	619	43°02'608"	21°24'746"
Prolom Spa 2	24	28	326	24	65	599	43°02'615"	21°24'393"
Prolom Spa 3	21	25	281	3.7	95	629	43°02'804"	21°24'025"
Prolom Spa 4	27	40	327	7.5	88	569	43°02'769"	21°23'656"
Prolom Spa 5	37	51	322	BDL	54	548	43°02'671"	21°23'398"
Lukovska Spa 1	16	20	411	1.1	20	774	43°09'867"	21°01'701"
Lukovska Spa 2	16	24	342	2.4	26	712	43°09'946"	21°01'990"
Lukovska Spa 3	9	15	263	1.4	20	692	43°09'986"	21°02'283"
Lukovska Spa 4	25	41	527	10	44	677	43°09'966"	21°02'487"
Lukovska Spa 5	26	53	700	0.3	53	684	43°10'143"	21°02'613"

BDL – below detection limit

The results of radon exhalation rate measurements are presented in Table 3. Descriptive statistics of radon exhalation rate $\mathbf{E_{Rn}}$ with the mean value of 50 mBq kg^{-1} h^{-1} is presented in Table 2.

According to the Shapiro-Wilk test the specific activity of ^{226}Ra and radon exhalation rate are consistent with a normal distribution (*Sig* = 0.786 and *Sig* = 0.376, respectively). A moderate correlation between radon exhalation rate and ^{226}Ra was found (Pearson correlation coefficient was 0.485). This correlation is illustrated in Fig. 4.

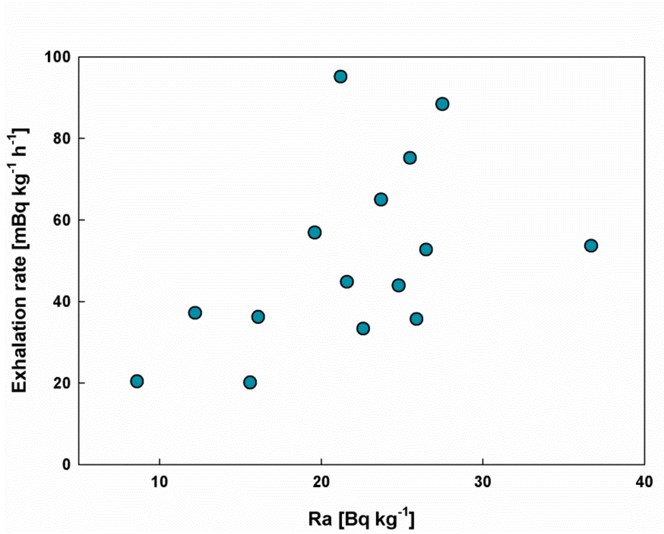

Fig. 4 Correlation between radon exhalation rate and ^{226}Ra

A certain difference in gamma dose rate measurements or in the values of specific activities of radionuclides could be explained by different geological units that exist in each of the spas (Fig.1). No correlation (Pearson correlation coefficient, r = -0.066) was found between calculated dose rate from radionuclides in soil and measured ambient gamma dose equivalent rate. A reason for non-existence of correlation could be treatment of soil samples before gamma spectrometry (grain size, humidity, density) [32]. Avdić et al. [4] reported stronger correlation between calculated and measured gamma dose rates if survey meter was placed directly on the ground. Besides, dose calculated from radioactivity in soil provide representative value of relatively small soil volume in comparison to measured dose which is contributed by much larger inhomogeneous volumes of soil [33, 34]. Measured values of ADER (1m above ground) are larger in comparison to calculated dose rate from soil radioactivity, which is to be expected given that ADER includes dose originating from cosmic radiation, self-effect of measuring device and contribution from ^{137}Cs [35]. Some other factors like weathering processes, vegetation cover, the soil moisture, season of measurements, etc. could significantly affect obtained results.

However, some authors reported strong correlation between calculated and measured gamma dose rates (r = 0.830), but they noted highly non-uniform distribution of radionuclides in small area due to localized mineralization [33]. Other studies reported the highest dose rates on sites with underlying acid intrusive background and the lowest dose rates recorded in the Quaternary geological background [36, 6]. In a study of relationship between dose rates and geological formations, authors pointed out that granitic origins give the highest terrestrial gamma dose rate while sandstone, sand clay and shale give the lowest dose rate [37]. Study conducted in Macedonia pointed out higher ambient dose equivalent rates in areas of volcanic geological origin than in areas of sedimentary origin [38]. A survey conducted in Budapest showed that results of ambient gamma dose equivalent rates were related to regional scale tectonic zones and surface sedimentary lithological conditions [39].

4. Conclusion

Specific activities of radionuclides in soil, radon exhalation rates, and ADER were measured in three spas in Southern Serbia. Values of measured ambient dose equivalent rates ranged from 73-170 nSv h^{-1} (with a mean value of 119 nSv h^{-1}). This is good approach to establish baseline background radiation, but it is only one reliable pattern for gamma radiation mapping of an area. Also, a larger number of measurements on diverse sites should be included. Additionally, an appropriate method for radiological risk assessment is to measure soil radioactivity. The calculated average specific activities (± standard deviations) of ^{226}Ra, ^{232}Th and ^{40}K in soils are 22±7, 31 ±11 and 420±128 Bq kg^{-1}, respectively. Results are comparable to other areas in Serbia. Further investigation found that radon exhalation rate from soil and specific activity of ^{226}Ra were moderately correlated (r = 0.485). Although this study has shown that no evidence can lead to a direct relationship between measured quantities and geology, an effort has been done to find a possible dependence. More various measurements need to be performed in order to establish a relation between geology features and a way of its radioactive appearance (manifestation) in environment.

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