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Field Experience with Soil Gas Mapping Using Japanese Passive Radon/Thoron Discriminative Detectors for Comparing High and Low Radiation Areas in Serbia (Balkan Region)

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Radon/Thoron/Natural radioactivity/Niška Banja/Obrenovac.

Based on results of fieldwork in the Balkan Region of Serbia from 2005 to 2007, soil gas radon and thoron concentrations as well as gamma dose rates were measured. Campaigns were conducted in two different geological regions: Niška Banja, considered a high natural radiation area, and Obrenovac around the TentB Thermal Power Plant (TPP), a low natural radiation area. Radon and thoron gas measurements were made by using two types of Japanese passive radon/thoron detectors, which included GPS data and gamma dose rates. The concentrations of soil radon gas in Niška Banja ranged from 1.8 to 161.1 kBq m^{-3}, whereas the concentrations for soil thoron gas ranged from 0.9 to 23.5 kBq m^{-3}. The gamma dose rates varied from 70 to 320 nGy h^{-1}. In the TentB area, radon concentration was found to range from 0.8 to 24.9 kBq m^{-3} and thoron from 0.6 to 1.9 kBq m^{-3}. The gamma dose rate ranged from 90 to 130 nGy h^{-1}. In addition, the natural radioactivity of the soil was investigated at the low background area. The radium and thorium contents in collected soil samples ranged from 23 to 58 and 33 to 67 Bq kg^{-1}, respectively. As a result of correlation analyses between the measured values, the highest correlation coefficient (R > 0.95) was found for thorium in the soil and the thoron gas concentration.

INTRODUCTION

According to a report by UNSCEAR[1)] the world mean annual effective radiation dose due to the inhalation of ^{222}Rn (radon), ^{220}Rn (thoron), and their decay products is estimated to be 1.26 mSv, with 1.15 mSv coming from ^{222}Rn and its progeny and 0.11 mSv from ^{220}Rn and its progeny.

In Niška Banja (located in the south-eastern part of Serbia; see the next section), several radiological surveys have been conducted. A preliminary (screening) survey was conducted for indoor radon/thoron.[2)] The soil-gas concentration ranges were from 63.7 to 1300 kBq m^{-3} and N.D. to 46 kBq m^{-3} for radon and thoron, respectively.

Niška Banja was identified as a high natural radiation area by this survey. Another detailed survey was conducted for radium in soil, radon in soil gas, and the gamma dose rate in a wider area of Niška Banja. The measurements show a significant correlation among radium in soil, radon in soil gas, and the gamma dose rate.[3)] Considering radon exposure and dose conversion factors such as that in ICRP 60[4)] and UNSCEAR, the estimated annual effective dose may exceed 50 mSv with a regional average of about 30 mSv.[5)] According to an earlier investigation, this value is close to the values for other high-level natural radioactivity areas (HLNRA).[6)]

However, the thoron concentration in soil gas was not

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measured in the whole area of Niška Banja, though previous studies have shown the existence of thoron in soil gas. Thus, the present study focused on soil thoron concentration in the Niška Banja area and its relationship between the gamma dose rate and soil radon concentration. As a control area (low natural radiation area), Obrenovac (TentB) (located in north-western Serbia, see the next section) was chosen.

In the Obrenovac (TentB) area, only a screening measurement was made and very low levels of radon and thoron concentration were reported.[7)] Radioactivity concentrations of U, Th, and ^{40}K were also tested[8)] for contamination of the soil and groundwater by thermal power plant waste, but no significant contamination was found.

The main objective of this paper is to compare the high and low background areas with a special attention to radon, thoron in soil gas, and their parent nuclide concentrations in soil, as well as the gamma dose rate measurement in the air at ground level.

Survey area

The present study was conducted at two places in Serbia. Niška Banja (180 km south-east of Belgrade) is regarded as an area with high-level natural radioactivity (HLNRA) and TentB (40 km south-west of Belgrade) as an area with low-level natural radioactivity (LLNRA).

a. Niška Banja

From a geological point of view, the town of Niška Banja is located in the quaternary alluvium of the River Nišava along the contact of Koritnik limestones and thick strata of travertine (spring sediments) end, which is also known as "bigar" in Serbian literature. This contact is also known as the "spa fault," which enabled the formation of a number of springs out of which the most important ones are Glavno Vrelo and Suva Banja.[9)]

b. Obrenovac (TentB)

The lithology of the Nikola Tesla TPP (TentB) surroundings is very simple. It is built up from quaternary river sediments consisting of loam and argillaceous sandy sediments. These lithological members usually lie across river terrace sediments consisting of marl clay, carbonic clay, diatomaceous earth, and sands of the Lower Pliocene (Pontian). The sediments are concordant usually without any tectonic disturbances. The only significant tectonic structure (south of TentB) is a fault that stretches in the E-NE direction close to the villages of Dren and Grabovac. The geological and hydrogeological profiles of the Obrenovac area show the presence of a main sandy gravel aquifer between two impermeable formations of shallow alluvial clay-like sediments and Miocenic marly clay.[10)]

MATERIALS AND METHODS

Measurements at Niška Banja were performed in November 2005 and at TentB in December 2006. Radon and thoron concentrations in soil gas were measured by using two types of passive radon-thoron discriminative detectors.[11,12)] These detectors were developed and evaluated at the National Institute of Radiological Sciences (NIRS) in Japan.

They can measure both radon and thoron concentrations. One type of the detector, called RADOPOT, was deployed at 56 points in Niška Banja, whereas for measurements around TentB at 27 sites a new type of the detector called RADUET was used. This is a remodeled version of the RADOPOT monitor, which is now made in Hungary and is commercially available. A comparison of these detectors showed that the differences between the results do not exceed 10%.[13)] Construction of these low cost detectors is simple, so the monitors have been widely used for radon and thoron surveys throughout the world.

This is the first time for RADOPOT and RADUET detectors to be used to measure radon and thoron concentrations in soil gas. The influence of humidity on radon/thoron measurement was tested at the NIRS and no significant influence (about 10%) was found.[14)] The typical error on the radon/thoron concentration measurement was assumed to be 20–25%,[15)] and therefore the humidity had only a small effect on the results.

Installation of the detectors at the survey areas was done as follows. Holes were drilled in the soil to a depth of about 80 cm depending on soil hardness. A detector was hung in the hole on a 70-cm stick and then the hole was covered by a polyethylene bag, which was pressed with an appropriate tile (Fig. 1).

Detectors were exposed for 4 days for Niška Banja and

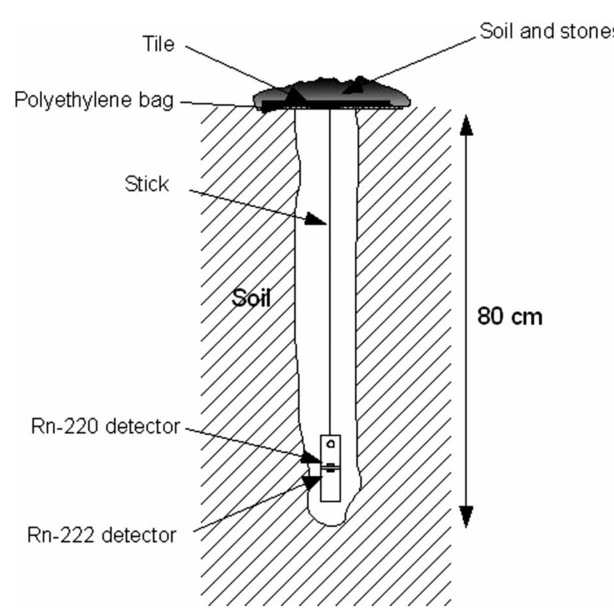


Fig. 1. Schematic of the measurement technique for radon/thoron in soil gas.

7 days for TentB, according to the expected radon concentration in the area. The CR-39 plates were etched for 6 h in 6.25 M NaOH solution at 90°C.[16)]

The measurements were performed in a grid pattern (with GPS coordinates) over the entire town of Niška Banja and around TentB, using a map of scale 1:25 000.

In the same locations, the gamma dose rate was measured using a scintillation counter. The measurements were performed 90–100 cm above ground level. Five parallel measurements (counting time: 100 s each) were conducted at every point. GPS coordinates were noted for every measuring point and later were used for mapping of the gamma dose rate and soil radon as well as the thoron concentration.

In addition, soil samples were collected from most of the measurement points in the TentB area to determine the concentrations of the natural radionuclides ^{40}K, ^{226}Ra, ^{228}Th, and ^{137}Cs. The samples were taken from a depth of about 50 cm and measured using a low-background NaI or HpGE detector. Samples were prepared in a plastic cylinder 9 cm in diameter and 4 cm high, closed, and aged for 2 to 3 weeks. The detector was characterized by Canberra, and an efficiency calibration made by using LABSOCS software (Canberra) was used for coincident summing corrections.

RESULTS

A total of 83 detectors were deployed for the measurement. Although detectors were deployed at 56 and 27 sites for Niška Banja and TentB, respectively, the concentration of soil thoron gas was not detectable at 6 sites. Moreover, data were excluded from further analysis if the measurement error exceeded the measurement value (16 cases). Results for concentrations of radon and thoron and for the gamma dose rates in both places are presented in Table 1. The Niška Banja to TentB ratios were calculated and are given as integers. The median value of radon concentration was 13.0 and 2.9 kBq m^{-3} in Niška Banja and TentB, respectively. The average radon concentration at each site was 26.5 ± 34 and 6.6 ± 7 kBq m^{-3}, respectively. The maximum radon concentration was 161.1 and 24.9 kBq m^{-3} with geometric mean of 14.5 and 3.9 kBq m^{-3}. The concentration of thoron gas was found with a geometric mean of 6.9 and 1.4 kBq m^{-3}, and ranged from 0.9–23.5 kBq m^{-3} for Niška Banja and 0.6–1.9 kBq m^{-3} for TentB.

The average radon concentration in the Niška Banja area was 4 times higher than around TentB and the thoron concentration was 7 or more times higher. The thoron/radon ratio for Niška Banja was more than 0.4 and 0.2 for TentB. The ratio of the gamma dose rate between the two areas was around 1.0. The average values were similar: 117 ± 50 and 114 ± 11 nGy h^{-1} for Niška Banja and TentB, respectively. The ranges of measurement values are different and amount to 250 nGy h^{-1} for Niška Banja with a maximum of 320 nGy h^{-1} and a minimum of 70 nGy h^{-1}, and a range

Table 1. Results for radon, thoron, and the gamma rate at Niška Banja and TentB.

	Parameter \ Area	Niška Banja (NB)	TentB (TB)	NB/TB ratio
Average + SD†	^{222}Rn [Bq m^{-3}]	26 500 ± 34 000	6 600 ± 7 000	4
	^{220}Rn [Bq m^{-3}]	10 600 ± 5 200	1 600 ± 500	7
	Gamma [nGy h^{-1}]	117 ± 50	114 ± 11	1
	^{226}Ra [Bq kg^{-1}]	–	48 ± 7	–
	^{228}Th [Bq kg^{-1}]	–	60 ± 8	–
Median	^{222}Rn [Bq m^{-3}]	13 000	2 900	4
	^{220}Rn [Bq m^{-3}]	8 400	1 700	5
	Gamma [nGy h^{-1}]	100	109	1
	^{226}Ra [Bq kg^{-1}]	–	49	–
	^{228}Th [Bq kg^{-1}]	–	62	–
GM (GSD)‡	^{222}Rn [Bq m^{-3}]	14 500 (400)	3 900 (3)	4
	^{220}Rn [Bq m^{-3}]	6 900 (200)	1 400 (2)	5
	Gamma [nGy h^{-1}]	–	113 (1)	–
	^{226}Ra [Bq kg^{-1}]	–	47 (1)	–
	^{228}Th [Bq kg^{-1}]	–	59 (1)	–
Range (min–max)	^{222}Rn [Bq m^{-3}]	1 800–161 100	800–24 900	–
	^{220}Rn [Bq m^{-3}]	900–23 500	600–1 900	–
	Gamma [nGy h^{-1}]	70–320	90–130	–
	^{226}Ra [Bq kg^{-1}]	–	23–58	–
	^{228}Th [Bq kg^{-1}]	–	33–67	–
No. of measurements	^{222}Rn [Bq m^{-3}]	56	22	–
	^{220}Rn [Bq m^{-3}]	49	5	–
	Gamma [nGy h^{-1}]	57	23	–
	^{226}Ra [Bq kg^{-1}]	–	23	–
	^{228}Th [Bq kg^{-1}]	–	23	–

†SD – standard deviation
‡GM – geometric mean and GSD – geometric standard deviations (in parentheses)

of 40 nGy h^{-1} for TentB with a maximum of 130 nGy h^{-1} and a minimum of 90 nGy h^{-1}. These data suggest that the distribution of radioactivity in soil is heterogonous at Niška Banja (Figs. 2a, b, c) and homogenous for the TentB area (Figs. 2d, e, f).

As shown in the geographical distributions of the radon/gamma dose rate in Niška Banja, the area where the maximum value of radon concentration was found closely corresponds to the area of maximum gamma dose rate. In other cases, the distribution is flat. If the thoron concentration was

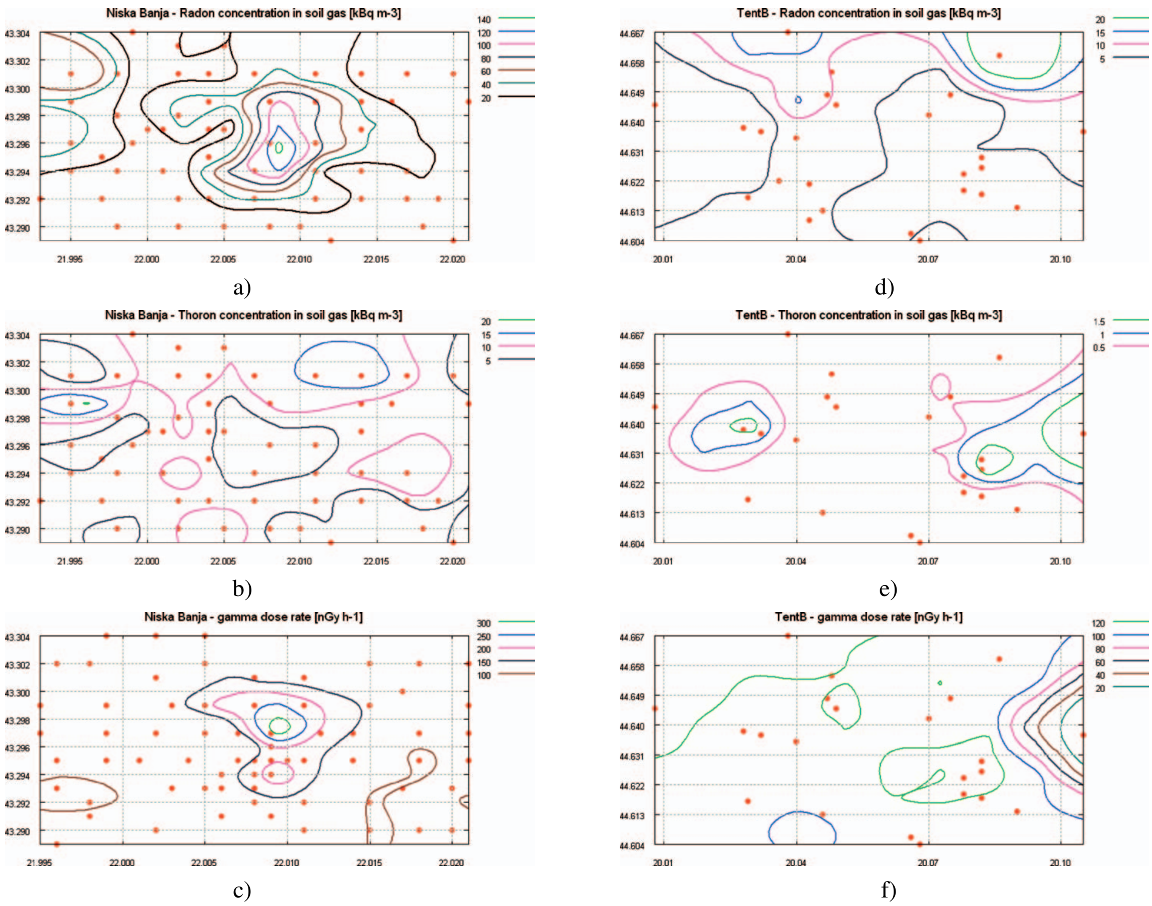


Fig. 2. Spatial distributions of the radon concentration in soil gas for a) Niška Banja and d) TentB, thoron concentrations at b) Niška Banja and e) TentB, and gamma dose rates at c) Niška Banja and f) TentB. The dots show the grid pattern of the measurements points.

lower than the error on its measurement, these data were not included in the spatial distribution of thoron for the TentB area (Fig. 2e). These data were also excluded from further analysis.

The averages of the gamma dose rate in both places were about 2 times higher than the world average (59 nGy h^{-1}). In neighboring countries UNSCEAR reported the following values: Slovenia, 56 nGy h^{-1}; Hungary, 61 nGy h^{-1}; Bulgaria, 70 nGy h^{-1}; and Romania, 54 nGy h^{-1}.

The soil samples were collected in the TentB area to determine the natural radioactive elements – ^{226}Ra, ^{228}Th, ^{40}K and ^{137}Cs. In a comparison between the Niška Banja and TentB results on soil thoron concentrations, it is interesting to analyze the data on ^{232}Th concentration in soil. The average ^{232}Th concentration in soil at TentB was 45 ± 6 Bq kg^{-1}, whereas a previous survey shows the average ^{232}Th concentration in Niška Banja to be nearly 40 Bq kg^{-1}. Nevertheless, the ^{220}Rn soil concentration in Niška Banja considerably exceeds the thoron soil concentration in the TentB region.

DISCUSSION

Natural radioactivity measurements were carried out at two areas in Serbia. The radon and thoron concentrations in soil in Niška Banja were higher than the concentrations found in soil in the TentB area, considered a “normal” background area. This can be explained by the differences in geological structure, content of natural radionuclides, and physical parameters of the soil.

Correlation analyses between measured values were performed and their results are presented in Table 2 for Niška Banja and Table 3 for TentB. By examining the Niška Banja results a strong correlation is observed between radon in soil and the gamma dose rate, with a correlation coefficient of 0.83. This result is close to the value obtained during an earlier investigation. Other calculated values presented in Table 2 shows no correlation (below 0.01).

An additional correlation analysis was done for TentB because this was the first investigation of natural radionuclides in soil. A spectrometric analysis for natural radionuclides in the soil samples resulted in average values of

Table 2. Correlation coefficients (R) between measured values for Niška Banja.

	^{222}Rn	Gamma	^{220}Rn
^{222}Rn in soil gas	1	–	–
Gamma dose rate	0.83	1	–
^{220}Rn in soil gas	< 0.01	< 0.01	1

48 ± 7 Bq kg^{-1} (with a range from 23 to 58 Bq kg^{-1}) for radium and 60 ± 8 Bq kg^{-1} (with a range from 33 to 67 Bq kg^{-1}) for thorium. In the soil samples taken at a depth of 50 cm, an artificial isotope of ^{137}Cs occurred with an average concentration of 1.7 ± 1.8 Bq kg^{-1}. The source of this isotope is unknown and further investigation is needed.

The highest value of the correlation coefficient ($R = 0.95$) was obtained for the thoron and thorium analyses (Table 3). Also, the good correlation ($R = 0.82$) was obtained between ^{228}Th and ^{226}Ra. Assuming the secular equilibrium between ^{238}U series and ^{232}Th series and their progenies, the correlation between radon and thoron should be good, but actually not ($R = -0.27$). This may be because soil radon concentration is affected by other factors than ^{226}Ra concentration (correlation coefficient between ^{226}Ra and ^{222}Rn is –0.50).

In contrast to Niška Banja, for TentB the parameter R showed no correlation between radon and the gamma dose rate ($R = 0.04$). Other measured values also showed no close correlation with each other. Fig. 3a, b shows the correlations between the radon and thoron concentrations for both areas. Although the correlation coefficient was calculated, the analysis provided no correlation (or negative correlation) between these parameters.

The average ratio of thoron/radon for Niška Banja was close to 40%, whereas the range was very wide—from 1% up to 266%. At TentB, this ratio did not exceed 12% with a minimum value of 3%, so thoron could be neglected.

The Shapiro-Wilk test for normality was performed and

Table 3. Correlation coefficients (R) between measured values for the TentB area.

	^{222}Rn	Gamma	^{226}Ra	^{228}Th	^{220}Rn
^{222}Rn in soil gas	1	–	–	–	–
Gamma dose rate	0.04	1	–	–	–
^{226}Ra in soil	–0.50	0.29	1	–	–
^{228}Th in soil	–0.31	0.31	0.82	1	–
^{220}Rn in soil gas	–0.27	–0.33	0.37	0.95	1

Table 4. Shapiro-Wilk normality test results.

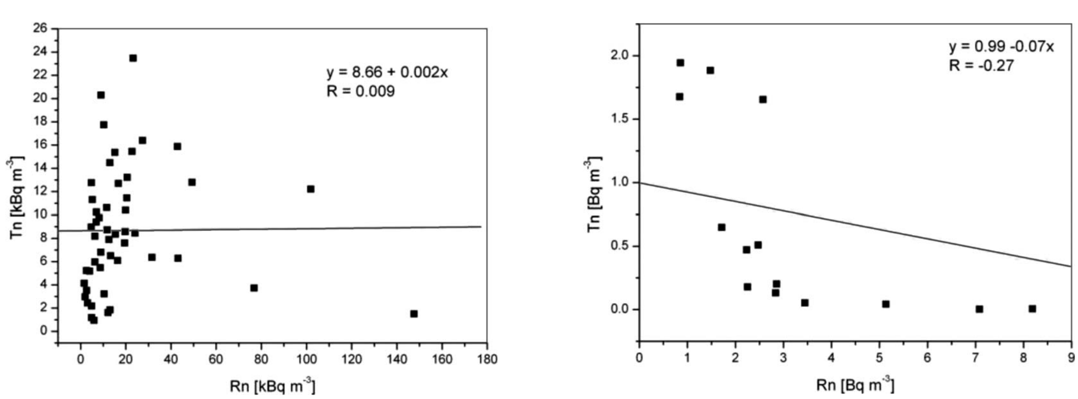
Parameter \ Area	Niška Banja		TentB	
	^{222}Rn	^{220}Rn	^{222}Rn	^{220}Rn
W	0.98	0.93	0.97	0.69
p	0.56	< 0.01	0.63	< 0.01

the results are reported in Table 4.

The W-statistic and p-value were computed, from which a statistical decision can be made by comparison with a level of significance. The logarithm of the radon concentrations clearly followed a lognormal distribution (Fig. 4a–b), whereas in the thoron case the parameter "p - calculated probability" were below the border value (assumption significance level of 0.05) for both areas.

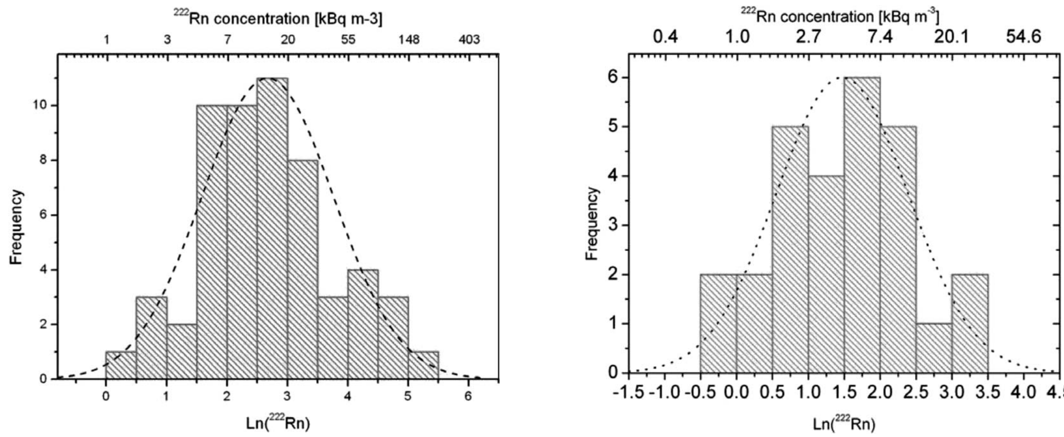
The simultaneous measurements of radon and thoron in soil gas at Niška Banja as well as at the TentB areas show that radon and thoron concentrations appear to be independent of each other. At some places in Niška Banja, the thoron concentration was 2 to 3 times higher than the radon concentration, whereas around TentB some places were found where thoron did not occur and the radon concentration was low (below 1.5 kBq m^{-3}).

Since the thoron problem has become worldwide, the



a) b)

Fig. 3. Correlation between soil radon and thoron concentrations for a) Niška Banja and b) TentB.



a) b)

Fig. 4. Lognormal distributions for radon at a) Niška Banja and b) TentB.

importance of radon–thoron discriminative measurements should be recognized especially in areas with complicated geological structure and where high concentrations of radium or thorium are found.

To explain the difference between thoron and thorium in the soil at each site (at the same depth, the concentration of thorium in Niška Banja was lower than that at TentB while the concentration of thoron was higher) the physical parameters of the soil (such as permeability and diffusion coefficients) as well as the identification of mineral need to be determined and further study is required.

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