# The Henryk Niewodniczański INSTITUTE OF NUCLEAR PHYSICS Polish Academy of Sciences

152 Radzikowskiego str., 31-342 Kraków, Poland

www.ifj.edu.pl/reports/2004.html Kraków, July 2004

**Report No 1946/AP** 

# PRELIMINARY RESULTS OF RADON AND THORON MEASUREMENTS IN SOUTH - EASTERN PART OF SERBIA AND MONTENEGRO

K. Kozak, J. Mazur, M. Janik, R. Haber

## Abstract

The results of measurements of radon and thoron concentrations in soil gas and in waters as well as radon exhalation rate, gamma dose rate and sampling soil for gamma spectroscopy measurements are reported.

Extremely high radon ( $^{222}$ Rn) concentrations were observed in soil gas (> 2000 kBq/m<sup>3</sup>) in Niška Banja town. Very high values of radon exhalation rates (1,5 Bq/m<sup>2</sup>s) as well as radon concentration in water samples (> 500 Bq/l) were also found. The next control measurements and monitoring of radon risk are planned for full recognition and understanding of that unusual phenomenon.

A survey on chosen areas in Serbia and Montenegro was done in June 2004. The first area (Niška Banja Spa) is located in the South-Eastern Serbia. The other one is Obrenovac, near Belgrade. This field work was organized and headed by dr Zora S. Zunic from ECE Laboratory (VINCA Institute of Nuclear Sciences, Belgrade).

## **INTRODUCTION**

Within the frame of bilateral state scientific cooperation agreement between Slovenia and Serbia and Montenegro, and scientific cooperation agreement between Institute of Nuclear Sciences "Vinča" and The Henryk Niewodniczanski Institute of Nuclear Physics Polish Academy of Sciences Krakow (IFJ PAN), the field work was carried out in two locations in Serbia (see Fig. 1), from June 11 to June 18, 2004.

- a) Niška Banja, Spa from June 11 to June 16, 2004;
- b) Obrenovac from June 17 to June 18, 2004.

The Natural Radioactivity Laboratory IFJ PAN, headed by dr Krzysztof Kozak is a subcontractor of **INTAILRISK** project within the frame of 5 UE Programme.



Fig. 1. Location of measurement sites in Serbia and Montenegro

The researchers form four institutions took part in the field work, dealing with several categories of measurements and using their own equipment. List of participants are presented in Table 1.

INSTITUTION	TEAM
VINČA Institute of Nuclear Sciences P.O. Box 522, 11001 Belgrade SERBIA and MONTENEGRO	<ol> <li>Dr Zora Zunic         <ul> <li>head of ECE Laboratory,</li> </ul> </li> <li>Igor Celikovic;</li> <li>Predrag Ujic.</li> <li>Violeta Evtov</li> <li>Bogdan Radulovic</li> </ol>
The Henryk Niewodniczański INSTITUTE OF NUCLEAR PHYSICS, Polish Academy of Sciences; Department. of Environmental and Radiation Transport Physics; Natural Radioactivity Laboratory (NRL); 31-342 Kraków, Radzikowskiego 152, POLAND	<ul> <li>6. Dr Krzysztof Kozak <ul> <li>head of the NRL, IFJ PAN</li> </ul> </li> <li>7. M.Sc. Jadwiga Mazur;</li> <li>8. M.Sc. Mirosław Janik;</li> <li>9.Tech. Ryszard Haber.</li> </ul>
JOŽEF STEFAN INSTITUTE Department of Physical and Organic Chemistry, Radon Centre Jamova 39, 1000 Ljubljana SLOVENIA	10. Dr Janja Vaupotič 11. Assoc. Prof. Ivan Kobal
UNIVERSITY OF KOSOVSKA MITROVICA Faculty of Natural Sciences Kosovska Mitrovica SERBIA and MONTENEGRO	12. Dr Gordana Krstic 13. Prof. Bajram Jakupi

 Table 1. List of participants.

The equipment used by the team of Natural Radioactivity Laboratory IFJ PAN is listed in Table 2 which includes also types and total number of performed measurements.

TYPE OF MEASUREMENT	UNIT	EQUIPMENT	Number of measurements
Radon concentration in soil	[kBq/m <sup>3</sup> ]	<ul> <li>Professional Radon Monitor AlphaGUARD PQ2000 PRO (Genitron, GmbH)</li> <li>Soil Probe</li> <li>AlphaPUMP with a set of pipes</li> <li>Drill</li> </ul>	8
Thoron concentration in soil	[kBq/m³]	<ul> <li>Professional Radon Monitor AlphaGUARD PQ2000 PRO (Genitron, GmbH)</li> <li>Soil Probe</li> <li>AlphaPUMP with a set of pipes</li> <li>Drill</li> </ul>	8
Radon concentration in water	[Bq/dm <sup>3</sup> ]	<ul> <li>Professional Radon Monitor AlphaGUARD PQ2000 PRO (Genitron, GmbH)</li> <li>AquaKIT (Genitron, GmBH )</li> <li>AlphaPUMP with a set of pipes</li> </ul>	7
Effective gamma dose rate	[nSv/h]	<ul> <li>Professional Radon Monitor AlphaGUARD PQ2000 PRO + GM Tube (Genitron, GmbH)</li> </ul>	9
Radon exhalation rate	[mBq/m <sup>2</sup> ·s]	<ul> <li>Professional Radon Monitor AlphaGUARD PQ2000 PRO (Genitron, GmbH)</li> <li>Exhalation Box with special Svagelock valves</li> <li>AlphaPUMP with a set of pipes</li> </ul>	8
Soil permeability	[m <sup>2</sup> ]	<ul> <li>Pressure Gauge VDPT-10S</li> <li>Soil Probe calibrated in Natural Radioactivity Laboratory IFJ PAN</li> <li>AlphaPUMP with a set of pipes</li> <li>Drill</li> </ul>	8
Meteorological parameters T-temperature, P-pressure, h-humidity	[°C] [Pa] [%]	<ul> <li>Professional Radon Monitor AlphaGUARD PQ2000 PRO (Genitron, GmbH)</li> </ul>	9
SAMPLING for Gamma Spectrometry in Natural Radioactivity Laboratory, IFJ PAN	[Bq/kg]	• Low-background gamma spectrometers with sodium-iodine detector NaI(Tl)	15

**Table 2.** Type of measurements, units, list of equipment and total number of measurements.

## **METHODS OF MEASUREMENTS**

The methods and equipment described below are applied for routine measurements carried out in our Laboratory.

#### Radon and thoron concentration in soil

The measurement setup consists of the AlphaGUARD PQ2000 PRO radon monitor, a soil gas probe and a pump (AlphaPUMP). A hole one-meter deep was drilled and the soil gas was pumped out into the ionization chamber of the AlphaGUARD (see Fig. 2). Both isotopes (<sup>222</sup>Rn and <sup>220</sup>Rn) were measured in this way. The average value of about thirty single, 1- minute measurements recorded by AlphaGUARD PQ2000 PRO was carried out. In order to determine only radon <sup>222</sup>Rn concentration, the ionization chamber was kept closed tightly after filling it with soil gas for about 10 minutes – time needed for thoron (<sup>220</sup>Rn) to decay. The concentration of thoron was determined as a difference between the first and the second measurement.

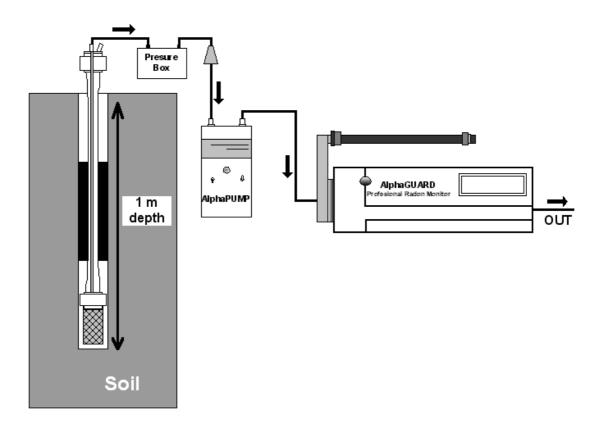


Fig. 2. Schematic set-up for radon and thoron in soil gas measurements .

## Radon exhalation rate

This parameter was measured "in situ" using the Exhalation Box with special valves (Swagelock type), a pump and the AlphaGUARD PQ 2000 PRO radon monitor which registers the increasing radon concentration inside the Exhalation Box (see Fig. 3). The value of exhalation rate has been determined from the slope of linear fit to the experimental data.

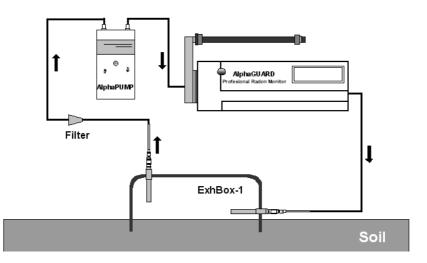


Fig. 3. Schematic set-up for radon exhalation rate measurements.

### Radon concentration in water

The professional radon monitor AlphaGUARD PQ2000 PRO with the additional equipment AquaKIT (a set of special glass vessels and pipes) was used for the measurements of radon concentration in water. Fig. 4 shows the set-up for this type of radon measurements. More details of the method can be found in Ref. [1].

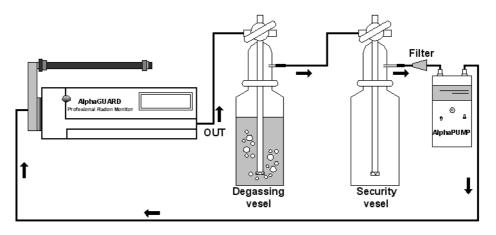


Fig. 4. Schematic set-up for measurements of radon in water.

#### Effective gamma dose rate

Effective gamma dose rate was measured using the professional radon monitor AlphaGUARD PQ2000 PRO with GM Tube. The registration of gamma dose rate was performed at 0 - 20 cm above ground level. Typical time of the measurement was 2 - 3 hours.

# Soil permeability

The measurement setup consisted of the Pressure Gauge VDPT-10S (range  $0\div10$  kPa), a rotameter ( $0\div1.0$  dm<sup>3</sup>/min), a soil gas probe and a pump (AlphaPUMP). A one-meter deep hole was drilled and the air was pumped into the hole (see Fig.5). In this way, the pressure difference and a flow rate could be measured. The modified equation from Fick's law is used to calculate the permeability:

$$k = \mu \frac{1}{W} \frac{Q}{dP}$$

where:

k – permeability [m<sup>2</sup>],  $\mu$  – dynamic viscosity [Pa·s], W – shape coefficient for soil probe [m], Q – flow rate [m<sup>3</sup>/s], dP – pressure difference [Pa].

The shape coefficient W is a semi-empirical parameter determined in our calibration device. It depends on flow rate and is different for different soil probe designs.

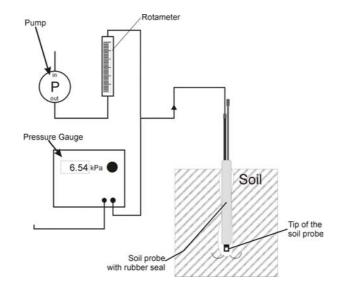


Fig. 5. Schematic setup for soil permeability measurement.

### Meteorological parameters

The meteorological parameters (air temperature, pressure and humidity) were registered by the radon monitor AlphaGUARD PQ2000 PRO during the measurements of all mentioned quantities.

#### Sampling for gamma spectrometry measurements

The soil samples were collected from the study area for the determination of concentrations of natural radioactive elements  $-{}^{226}$ Ra,  ${}^{232}$ Th and  ${}^{40}$ K. They were taken from the depth of 0.6  $\div$  1.0 m while drilling holes for radon measurements in soil. The concentrations of natural radio-isotopes are determined in laboratory by means of scintillation spectrometry with 3"x3" NaI(Tl) detector.

# RESULTS

The results of all measurements performed by Natural Radioactivity Laboratory IFJ PAN are gathered in Table 3 and Table 4. Table 3 contains the results of "soil measurements" and meteorological conditions registered during these measurements. The results of radon concentrations in water are presented in Table 4. The measurement sites are coded in both tables. The codes 1 - 6 in Table 3 apply to soils near detached houses in Niška Banja and the codes 7 - 8 concern the sites near Obrenovac.

The extremely high radon (<sup>222</sup>Rn) concentrations are observed in soil gas in Niška Banja town, ranging from  $64 \text{ kBq/m}^3$  to above  $2\ 000\ \text{kBq/m}^3$ . The latter value is <u>out of AlphaGUARD PQ 2000 PRO upper detection limit</u>. Very high values of radon exhalation rates (1,5 Bq/m<sup>2</sup>s – code 5 in Table 3) are also noticed. The contribution of thoron concentration amounts to 62% of that of radon <sup>222</sup>Rn.

Water samples were collected from natural sources (codes 9 - 13 in Table 4) and from the water-supply system (codes 14, 15 in Table 4). The sample 9 was taken from the thermal source in Niška Banja. Because it was impossible to take a sample from the proper source it was taken directly from an "open" stream which was flowing out of the hidden source. Such method of water sampling may cause the significant underestimating of the calculated radon concentration due to radon escape from a sample.

CODE of measure-	DATE	AIR TEMPERATURE	AIR PRESSURE	AIR HUMIDITY	EXHALATION RATE	CONCENTE SO	IL	GAMMA DOSE	SOIL PERMEABILITY
ment	DATE	[°C]	[hPa]	[%]		RADON	THORON	RATE	
place		(min ÷ max)	(min ÷ max)	(min ÷ max)	[mBq/m <sup>2</sup> ·s]	[kBq/m³]	[kBq/m³]	[nSv/h]	[m <sup>2</sup> ]
1	1 12-06-04	32.9	990.5	50	30 ± 6	63.7 ± 2.2	$20 \pm 4$	146	$1.11 \cdot 10^{-12} \pm 3.41 \cdot 10^{-14}$
1		$(29.5 \div 36.0)$	(990.4 ÷ 990.6)	$(41 \div 60)$					
C	2 12-06-04	30.4	986.4	52	96 ± 18	$77.5\pm2.6$	5 ± 4	152	$2.77{\cdot}10^{-12}\pm1.70{\cdot}10^{-13}$
Δ		$(27.5 \div 34.8)$	$(986.2 \div 986.4)$	$(46 \div 56)$					
3	13-06-04	29.7	989.3	56	$450\pm71$	> 2000	not	276	$2.13 \cdot 10^{-12} \pm 1.01 \cdot 10^{-13}$
	13-00-04	$(27.3 \div 31.6)$	$(989.2 \div 989.5)$	$(48 \div 62)$			measured	270	
4	14-06-04	24.2	996.1	75	$671 \pm 86$	$1300\pm45$	0 ± 62	194	$4.51 \cdot 10^{-12} \pm 2.89 \cdot 10^{-12}$
4	14-00-04	$(20.6 \div 28.4)$	(995.9 ÷ 996.3)	$(66 \div 84)$	071 ± 80				
5	14-06-04	25.8	992.4	66	$1542 \pm 354$	980 ± 33	$20 \pm 50$	203	$2.67 \cdot 10^{-12} \pm 4.38 \cdot 10^{-13}$
	14-00-04	$(24.3 \div 27.0)$	$(992.3 \div 992.4)$	$(62 \div 70)$	1342 ± 334				2.07.10 ±4.36.10
6	6 15-06-04	24.0	993.8	65	57.9 ± 11.9	$74.3\pm2.4$	$46 \pm 5$	128	$1.41 \cdot 10^{-12} \pm 2.01 \cdot 10^{-13}$
0		$(23.5 \div 24.6)$	(993.6 ÷ 994.1)	$(62 \div 70)$					1.41.10 ± 2.01.10
7	7 17-06-04	22.6	1003.0	42	Not	0.099	not	89	$1.77 \cdot 10^{-14} \pm 8.85 \cdot 10^{-15}$
/ 1/-		$(21.4 \div 24.8)$	(1003÷1004)	$(39 \div 46)$	measurable	measurable $\pm 0.033$ measured	09	1.77.10 ± 0,03.10	
0	8 18-06-04	23.5	998.7	54	9 ± 4	0.691 ± 0.10	not	135	$3.35 \cdot 10^{-14} \pm 1.36 \cdot 10^{-14}$
0 10		$(22.5 \div 25.3)$	$(998.5 \div 998.8)$	$(48 \div 61)$			measured		5.55.10 ± 1.50.10

Table 3. Results of measurements performed by Natural Radioactivity Laboratory IFJ PAN during field work in Serbia and Montenegro.

Sample CODE	DATE	RADON IN WATER [Bq/dm <sup>3</sup> ]
9	13-06-04	$23 \pm 2$
10	13-06-04	$460 \pm 17$
11	13-06-04	$0.2 \pm 0.4$
12	14-06-04	$571 \pm 21$
13	14-06-04	$507 \pm 19$
14	15-06-04	$0.5 \pm 1.6$
15	15-06-04	$3.0\pm0.5$

**Table 4.** Results of measurements performed by Natural Radioactivity Laboratory IFJ PAN during field work in Serbia and Montenegro.

#### **CONCLUSIONS**

The team of Natural Radioactivity Laboratory IFJ PAN (Kraków) took part in the field work organized by the Vinča Institute of Nuclear Sciences (Belgrade) and performed the first measurements of radon and thoron concentrations in soil, of radon exhalation rate and of radon concentration in water on the chosen research areas – Niška Banja Spa in South-Eastern Serbia and Obrenovac near Belgrade. Teams from other institutions (see Table 1) made other types of measurements – e.g indoor radon and radon progeny concentrations.

The presented preliminary results of radon and thoron concentration measurements in one of investigated areas (Niška Banja) show extremely high levels. This region is planned to be investigated thoroughly in near future. The research will be upgraded with geological identifications.

All results, including those of the next set of field measurements, will be published.

We gratefully acknowledge Dr Zora S. Zunic for the organization and inviting us to take part in the field work in Serbia and Montenegro.

We also thank our colleague Tadeusz Zdziarski for help in performing the measurements – especially for design and construction of the soil probe.

# LITERATURA.

[1] E. Kochowska, J. Mazur, K. Kozak, M. Janik, "Radon in Well Waters in the Krakow Area", Isotopes in Environmental and Health Studies **40** (2004) - in print.