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METHODS OF RADON MEASUREMENT

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Abstract. *Several important international scientific organizations have designated radon as a carcinogenic and serious health problem. As a chemically inert gas, it is easily released from soil, building materials, and water, to emanate to the atmosphere. Since 1992, Laboratory for Nuclear Physics of the Department of Physics, Faculty of Sciences in Novi Sad has been involved in measurements of radon concentration in air, using several different techniques. Last year, systematic radon measurements in drinking waters began, too. The work presented here gives a survey and discussion of the results of the both series of measurements.*

Key words: *radon activity concentration, charcoal canisters, gamma-spectrometry, soil radioactivity, emanation, CR39 track detectors, alpha spectrometer.*

INTRODUCTION

Research carried out in recent decades has shown that, under normal conditions, more than 70% of a total annual radioactive dose received by people originates from natural sources of ionizing radiation, whereby 40% is due to inhalation and ingestion of natural radioactive gas radon ^{222}Rn and its decay products (Fig. 1). Exposure to radon via inhalation in closed rooms is the cause of about 10% of all deaths from lung cancer. Changes at the cellular and molecular levels are significantly more pronounced in early stages of life [1, 2].

Radon is a natural inert radioactive tasteless and odorless gas, whose density is 7.5 times higher than that of air. It dissolves in water and can readily diffuse with gases and water vapor, thus building up significant concentrations. The physical half-life of radon is 3.825 days and half-elimination time from lungs 30 min. Radon ^{222}Rn, which is the daughter of uranium ^{238}U, represents the most important radon isotope. Decay of the radon nucleus ^{222}Rn yields short-living daughters: polonium ^{218}Po, lead ^{214}Pb and bismuth ^{214}Bi.

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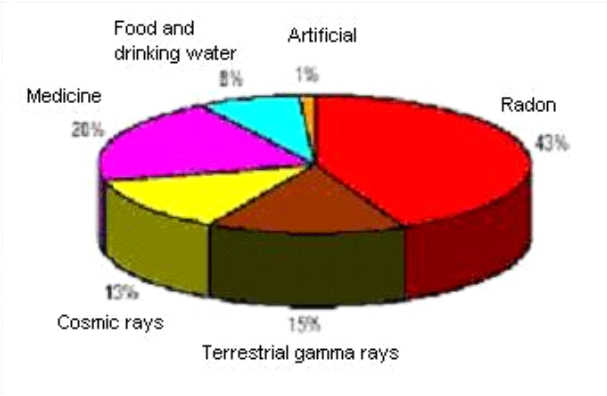

Fig. 1. Estimation of total annual dose received by people

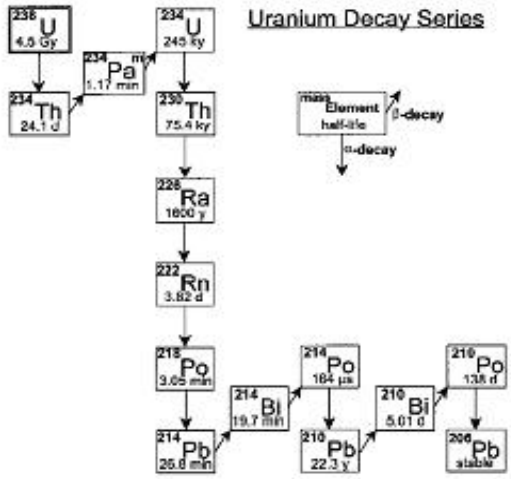

Figure 2. Decay scheme of ^{238}U

RADIATION HEALTH RISK AND ALARA PRINCIPLE

According to the recommendations of the International Commission on Radiological Protection (ICRP) from 1994, the civil engineering standard for building new apartments and houses is 100 Bq/m^3 radon as an average annual level, 200 Bq/m^3 for the recommendation of inexpensive remediation measures, and 600 Bq/m^3 for the recommendation of expensive remediation measures. Namely, even in the case of observing these recommendations and norms, ALARA (As Low As Reasonably Achievable) principle applies [3, 4].

According to the Regulation on the Limits of Exposure to Ionizing Radiation (Official Journal of SRJ No.32/1998, Art. 16), intervention levels for chronic exposure to radon in apartments is equal to one half of the mean annual concentration of 200 Bq/m^3 ^{222}Rn in

the air of newly-built apartments and 400 Bq/m^3 ^{222}Rn in the air of the already existing housing facilities. Hence, if the absolute abundance in parts of the lithosphere or geochemical processes, regionally or locally, concentrate natural radio-nuclides, then higher ionization levels are to be unavoidably tolerated.

The EPA recommendation for the upper limit of radon concentration in drinking water is 300 pCi/l, which amounts to 11.1 Bq/l. As our country has no regulations or recommendations on allowed concentration levels of radon radioactivity in drinking water, the obtained results are considered in view of the USA standards. The dominant source of radon in closed rooms up to the second floor of a building is the building soil (the soil beneath the housing object), whereas in the rooms on higher floors radon originates from the building materials. Significant factors that influence radon concentration that is accumulated in apartments include: soil characteristics (chemical composition, physical and hydro-mechanical properties, porosity among the first), quality of constructing (construction type, ventilation), as well as climatic changes and meteorological conditions.

MEASUREMENT METHOD

Since 1992, in the Laboratory of Nuclear Physics, Department of Physics in Novi Sad, measurements of radon activity concentration have been carried out in closed room, as well as of radon emanated from soil, using the method of adsorption on activated carbon. To the present, more than 200 measurements have been carried out with the aid of charcoal canisters (Fig. 3). Calibration and correction of the method was performed using EPA standards. Concentrations of radon activity were determined on the basis of the intensity of gamma-lines of short-living radon daughters ^{214}Bi and ^{214}Pb. Gamma-spectrometric measurements were carried out using a high-resolution high-purity germanium detector of nominal efficiency of 22%, placed in a protection chamber with 25-cm thick iron walls. Detector efficiency was determined via the ^{226}Ra reference source made by EPA.

Detector exposure lasted in average two days. Radon emanation from soil was measured using a steel probe dug in the earth to the depth of 70 cm with the upper end closed and a metal grid on which a charcoal canister was placed at the bottom end (Fig. 4). Since soil represents the dominant radon source, soil samples taken from the location of radon emanation, after the appropriate preparation, were subjected to measurement of radionuclides concentration by the gamma-spectrometric method.

With the aim of constructing a first radon map of Vojvodina, activity concentration has been measured using a CR39 trace detector on about 1,000 different locations in all 45 municipalities in the period December 2002 - March 2003. To cover all municipalities, the detectors were distributed to local physics teachers, who further distributed them among their pupils. CR39 trace detectors were placed in the ground-floor rooms, in living rooms or bedrooms, at a height of 1 m above the floor, far from water or heat source. The selected locations were in suburban and rural settlements with old type houses, having earth floors or poorly isolated concrete constructions. Measurements within the same scope were repeated in the winter-spring season of 2004 and 2005.

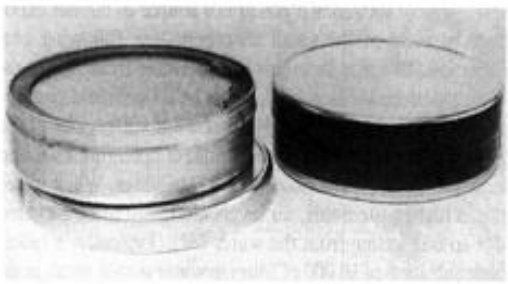

Fig. 3. Photographs of charcoal canisters made according to EPA standards

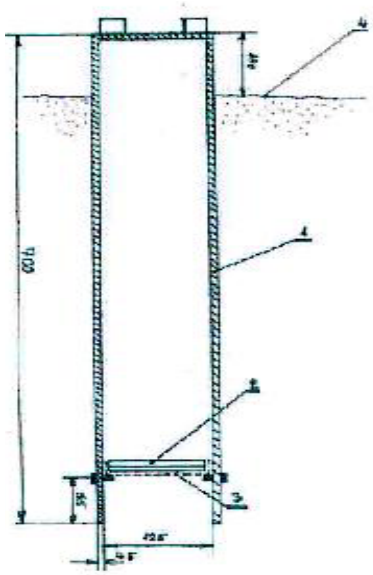

Fig. 4. Sketch of the probe for radon emanation measurement: 1 - steel tube, 2 – charcoal canister, 3 - metal net, 4 – soil surface layer

CR39 radon detectors represent plastic films with an area of 1 cm^2, 1 cm thick, and sensitive to traces of ionizing alpha particles [5]. During the exposure, these detectors were glued to the cover of a closed diffusion plastic chamber 5 cm high. The detector is sensitive to alpha radiation only, and its sensitivity equals 2.9 traces/(cm^3 kBqh/m^3). The detectors were etched in 25% solution of NaOH at a constant temperature of 90^0C during 4 hours. The traces were read and treated by RADOSYS 2000 electronic equipment (Fig. 5) in the Radosys Company, Hungary. This sophisticated equipment (Fig. 5) includes: RADOBATH 2000 (thermostated bath for chemical etching of traces on the detectors) and RADOMETER 2000 equipment for reading traces, with a B&W CCD camera and a compatible PC. Average exposure time of CR39 detectors was 90 days.

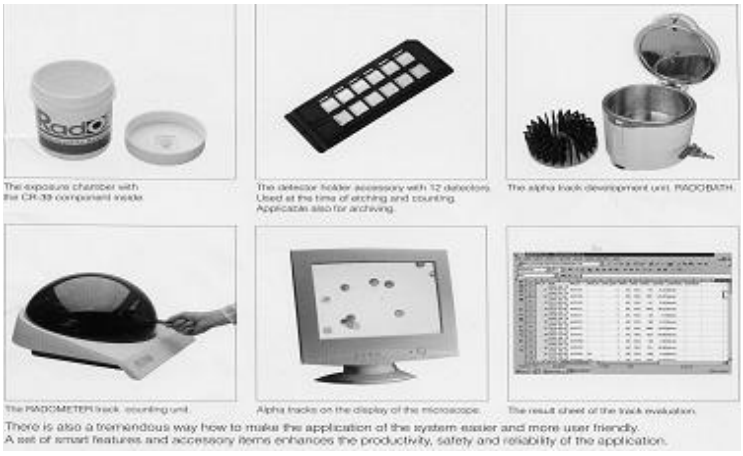

Fig. 5. CR39 radon detectors placed in plastic diffusion chambers with displayed procedures of developing and etching traces and RADOSYS 2000 electronic equipment.

Measurements of radon concentration in drinking water was carried out using a calibrated alpha spectrometer DURRIDGE ***RAD 7*** (Fig. 6), with special accessories for radon measurement in water. The detector converts alpha radiation directly to an electric signal and has the possibility of determining electronically the energy of each particle, which allows the identification of the isotopes (^{218}Po, ^{214}Po) produced by radiation, so it is possible to instantaneously distinguish between old and new radon, radon from thoron, and signal from noise.

Fig. 6. Measurement apparatus RAD 7

MEASUREMENT RESULTS

On the basis of the previous measurements of soil radioactivity, it was possible to determine mean, minimal, and maximal activity concentrations of the natural radio-nuclides ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K, as well as of the radionuclide ^{137}Cs (Table 1).

Table 1. Results of radioactivity measurements of soil [6]

Radionuclide	$\bar{A}_{mean}$ [Bq/kg]	A_{min} [Bq/kg]	A_{max} [Bq/kg]
^{137}Cs	12 ± 9	1.1	55.0
^{238}U	51 ± 9	24.0	72.0
^{226}Ra	39 ± 7	19.7	51.0
^{232}Th	53 ± 8	22.0	64.0
^{40}K	554 ± 92	238	730

The obtained values of radionuclide concentrations are within the range of usual radioactivity limits for agricultural soils. ^{137}Cs was detected in all soil samples, and the large span between minimal and maximal concentrations suggests an artificial origin of this radionuclide (accident at the Chernobyl nuclear power station in 1986). Since the half-life of ^{137}Cs is 30 years, this isotope will be long present in the Vojvodina environment.

On the given locations, apart from measuring soil radioactivity, measurement of radon emanation was also carried out. Table 2 gives mean values with standard deviation, as well as minimal and maximal concentrations of radon emanated from soil. As no correla-

tion has been established between activity concentration of ^{238}U – ^{226}Ra (Table 1) and intensity of radon emanation from soil (showing significant dispersion around the mean value), it can be concluded that this process depends on the rock disposition in the soil, soil porosity, and groundwater flows.

Table 2. Results of measuring radon emanated from soil

No. of measurements	A_{mean}[Bq/m^3]	σ(A)[Bq/m^3]	A_{min}[Bq/m^3]	A_{max}[Bq/m^3]
70	987	733	44	3247

In Fig. 7 it is shown the log-normal distribution of measurement results, which indicates that radon emanation from soil is a natural process. The distribution is shifted towards lower concentrations of radon activity, and the measured values are common for this region of Europe [7].

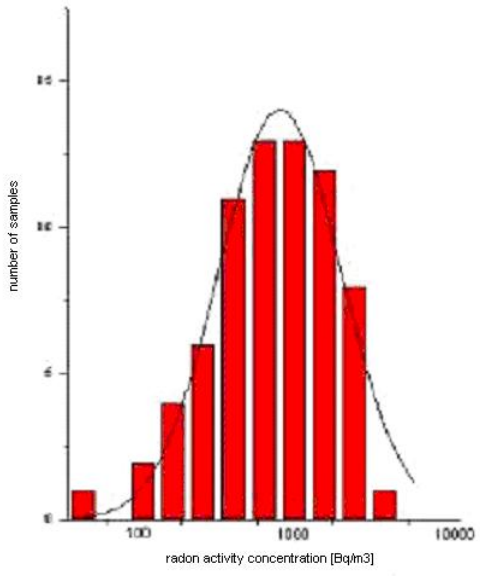

Fig. 7. Log-normal distribution of radon emanated from soil

The results of measuring the activity concentration of radon in closed rooms that have been carried out on about 200 different locations in the area of Novi Sad municipality in the period from 1992 to the present are shown in a tabular form (mean, minimal, and maximal measured radon concentration) in Table 3 and as the log-normal distribution in Fig. 8.

Table 3. Results of radon measurements in Novi Sad apartments by charcoal canister method

Radionuclide	A_{max} [Bq/m^3]	A_{min} [Bq/m^3]	A_{mean} [Bq/m^3]	$\sigma(A)$ [Bq/m^3]
^{222}Rn	391	2	50	69

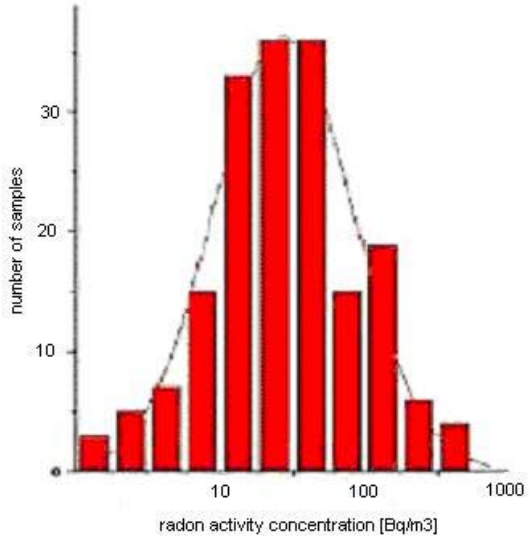

Fig. 8. Log-normal distribution of radon concentration in Novi Sad apartments measured by charcoal canister method

These values can be compared with the values of radon concentration obtained in the long-term measurements by the method with a CR39 detector for Novi Sad (Table 4). The values measured by this method are somewhat higher compared with those in Table 3, which can be explained in terms of weather conditions (mainly winter ones) and the location of the detectors mainly in suburban settlements in old ground-floor houses with poor floor isolation.

Measurements of radon concentration by CR39 detectors for 2005 are still in progress, whereas the results for 2003 and 2004 are presented in the form of radon maps in which for each territorial unit (municipality) is given the value of mean radon concentration in closed rooms (Figs. 9 and 10). Total number of measurements, mean, minimal and maximal indoor radon concentrations, as well as locations on which these values were obtained are given in Table 5 and Table 6.

Table 4. Results of measuring radon concentration in Novi Sad apartments by CR39 method

Municipality	A_{maen} [Bq/m^3]	$\sigma(A)$ [Bq/m^3]	n No. of meas.	A_{min} [Bq/m^3]	A_{max} [Bq/m^3]
Novi Sad	133	115	86	10	445

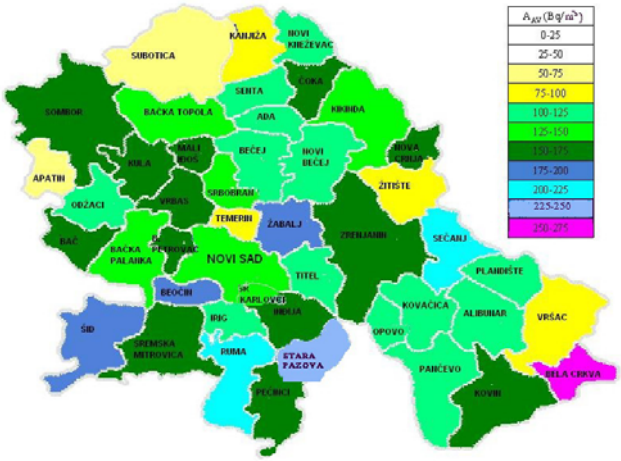

Fig. 9. Radon map of Vojvodina with the measurement results for 2003

Table 5. Results of radon map measurements for 2003

A_{mean} [Bq/m^3]	n No. of meas.	A_{min} [Bq/m^3]	Location	A_{max} [Bq/m^3]	Location
144	968	2	Ada	893	Zrenjanin

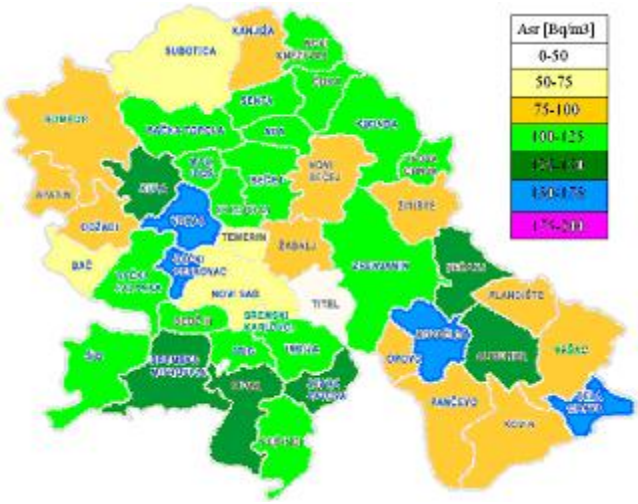

Fig. 10. Radon map of Vojvodina with the measurement results for 2004

Table 6. Results of radon map measurements for 2004

A_{mean} [Bq/m^3]	n No. of meas.	A min [Bq/m^3]	Location	A max [Bq/m^3]	Location
102	941	1	Plandište	599	Bela Crkva, Inđija

During 2004, radon concentration was measured in all bottled drinking waters that could be purchased in the market. Also, the same measurements were carried out for the water from the waterworks of Novi Sad and Backi Petrovac, to compare the results with those for bottled waters (Table 7).

Table 7. Results of radon measurements concentration in drinking waters

Water	Bottling Date	Measurement Date	Activity Concentr. (Bq/l)
KNJAZ MILOŠ 1.5 l, non-carbonated	12.8.2004.	16.8.2004.	<0.26
VRNJCI 1.5 l, carbonated	3.8.2004.	23.8.2004.	<0.11
AQUA VIVA 1.5 l, non-carbonated	14.7.2004.	23.8.2004.	<0.11
VODA VODA 0.5 l, non-carbonated	8.7.2004.	24.8.2004.	<0.16
ROSA 0.5 l, non-carbonated	27.7.2004.	24.8.2004.	<0.48
Water from Novi Sad waterworks		24.8.2004.	3.9±0.5
PI-VODA 1.5 l	25.8.2004.	26.8.2004.	2.1±0.7
DUBOKA 0.5 l, mineral	22.6.2004.	26.8.2004.	<0.5
BIVODA 1.5 l, carbonated	31.5.2004.	26.8.2004.	<0.11
KARADJORDJE 1.5 l, non-carbonated	29.6.2004.	26.8.2004.	<0.15
MIVELA 2 l, carbonated	19.7.2004.	26.8.2004.	<0.35
EVIAN 1.5 l, non-carbonated	19.5.2004.	26.8.2004.	<0.5
PROLOM VODA 1.5 l	18.8.2004.	27.8.2004.	2.8±0.7
EKO VODA 1.5 l, non-carbonated	12.6.2004.	27.8.2004.	<0.41
AQUA BIANCA		11.11.2004.	<0.16
SANPELLEGRINO 0.25 l, carbonated		6.12.2004.	0.15±0.07
Water from B. Petrovac waterworks		8.12.2004.	0.75±0.08
DUBOKA 0.33 l, carbonated	14.6.2004.	8.12.2004.	<0.11
VODA VODA 0.5 l, non-carbonated	1.10.2004.	8.12.2004.	<0.11
VLASINSKA ROSA 0.5 l, non-carb.	30.10.2004.	20.12.2004.	<0.33
MINAQUA 0.5 l, carbonated	25.10.2004.	20.12.2004.	<0.11
KNJAZ MILOŠ 1.5 l, carbonated	10.12.2004.	20.12.2004.	0.26±0.07
PROHOR 5 l, weakly carbonated	9.9.2004.	22.12.2004.	<0.15
FAST WATER 1.5 l, carbonated	23.1.2004.	22.12.2004.	<0.11
AQUA BELLA 0.33 l, carbonated	9.9.2004.	22.12.2004.	<0.1
AQUA HEBA 1.5 l, carbonated	30.7.2004.	16.8.2004.	<0.1
SKADARLIJSKA 1.5 l, carbonated	4.6.2004.	27.8.2004.	<0.1

The results above show that in view of radon concentration, having in mind the EPA recommendation for allowed radon level in drinking waters of 11.1 Bq/l, all bottled waters are safe for utilization.

CONCLUSION

Almost 13% of the results of the radon concentration measurement in apartments and houses of Vojvodina exceeded the limit value of 200 Bq/m^3, whereas 1% of measurements showed an extremely elevated radon concentration (above 400 Bq/m^3). The results are significantly higher than the expected ones (compared with previous results, Table 3),

which indicates that, in the plain areas too, although the ^{238}U concentration is within usual limits (Table 1), radon concentration may be elevated, probably because of the groundwater effects. Repeated measurements on the locations where radon activity concentration was above 200 Bq/m^3 showed a decreasing tendency in the current year (which can be seen from the radon maps for 2003 and 2004 in Figs. 9 and 10). The owners of these houses adopted the recommendations and technical advices for lowering radon concentration in closed rooms (better ventilation, concreting the floor and reinforcement of floor construction, utilization of isolating materials) given by the expert team from the Laboratory of Nuclear Physics of the Department of Physics Novi Sad.

Further measurements of radon activity concentrations in Vojvodina should be continued with the aim of finding out the areas with elevated values for this radionuclide and obtaining more complete data for the radon map of Vojvodina.

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METODE MERENJA RADONA

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Značajne međunarodne naučne organizacije su označile radon kao karcinogen i ozbiljan zdravstveni problem. Kao hemijski inertan gas, radon lako napušta zemljište, građevinski materijal i vodu i prelazi (emanira) u atmosferu. Laboratorija za nuklearnu fiziku, Departmana za fiziku na PMF-u u Novom Sadu se od 1992. godine bavi merenjem koncentracije radona u vazduhu i emaniranog iz zemljišta korišćenjem više različitih metoda. Prošle godine započeto je i merenje koncentracije radona u pijaćim vodama. U ovom radu dat je pregled rezultata dosadašnjih merenja uz diskusiju dobijenih rezultata.