RADIOECOLOGICAL PROBLEMS IN HOME BUILDING IN THE CITY OF NOVI SAD

S.Curcic[1)], I.Bikit[1)], M.Veskovic[1)], J.Slivka[1)], Lj.Conkic[1)], E.Varga[1)], N.Zikic-Todorovic[1)], D.Mrdja[1)]

[1)] Department of Physics, Faculty of Sciences, University of Novi Sad, Novi Sad, Serbia and Montenegro, tel:+381 21 455 318, fax:+381 21 455 318, e-mail: sofija@im.ns.ac.yu

Abstract

The city of Novi Sad lies on the Panonian low-land without elevate uranium levels in the surface soil. However the rich underground waters still can cause radon buildup problems. In order to examine this problems a series of measurements on building material, soil, radon emanation from soil and radon buildup in houses and flats were performed. The gamma spectroscopy measurements on building materials and soil resulted in moderated natural radioactivity (^{238}U, ^{226}Ra, ^{232}Th) levels with the order of magnitude 50 Bq/kg. The activity concentration of the radon emanated from soil was about 1000 Bq/m^3 with significant local variations. The radon concentrations were measured by charcoal absorbers and CR-39 track detectors, revealing 27% houses with radon above the 200 Bq/m^3 action level.

Introduction

The soil of Vojvodina may contain radioactive contaminants from a number of sources. First of all, these are the nuclear power plants in the South East Europe region that could contaminate this region through the release of radionuclides into the water. The use of phosphate fertilizers with high uranium concentration may also cause a gradual increase of the uranium series activity concentration in soil. The concentration of uranium and thorium in Earth's crust is in the range 1.1 – 10 ppm [1] for uranium and 10 ppm [2] for thorium. This corresponds to an activity concentration range of 13.5 – 123 Bq/kg for ^{238}U and 39.4 Bq/kg for ^{232}Th. Beside the daughters of natural radioactive series, long-lived natural isotope ^{40}K is also greatly presented in the soil.

Radon is a naturally occurring radioactive gas. It is a colorless and odorless noble gas, 7,5 times weighted than the air. Uranium ^{238}U decays through several different isotopes. When it reaches radium ^{226}Ra, it decays to radon ^{222}Rn and undergoes a change of state to a gas. Radon partially decays in material where it has been generated and partially moves rapidly by concentration-driven diffusion into the open air. Sources of radon in flats are: soil under and round the object, building's material and water and gas used in household. Indoor radon concentration depends on: ^{226}Ra content in soil and building materials (BM), moisture content in soil and BM, potential of diffusion in soil and BM, surface area and isolation quality of structures contacting with soil, building floor, air ventilation in the building, weather conditions and indoor-outdoor temperature and pressure differences [3].

The base of developing the public indoor radon politics is a definition of risks connected with the exposition of population to radon and its daughters in the living rooms. The actions that are conducted in order to reduce radon activity concentrations in flats and offices suppose the establishment of national recommendations, directives or laws about maximal permitted radon concentrations [4,5].

BM causing high concentration have been used in several countries. In some cases these are materials of natural origin (i.e., granite or alum shale concrete), and in other cases they are by-products from different industries (by-product gypsum, waste rock from mining etc.). Serbia and Montenegro has advisory reference levels for internal BM: ^{226}Ra< 200 Bq/kg; ^{232}Th<300 Bq/kg; ^{40}K<3000 Bq/kg; all antropogenic radionuclides< 4000 Bq/kg. Maximum permitted annual indoor dose due to gamma radiation from BM is 1 mSv, which corresponds to the following activity index :

$$^{226}\text{Ra}/200 + {}^{232}\text{Th}/300 + {}^{40}\text{K}/3000 + \text{all antropogenic radionuclides}/4000 < 1 \qquad (1)$$

Methods

The soil and building materials samples were dried at 105^{o}C to constant mass and transferred to sample holders. Due to urgency to cary out radioactivity measurements ther was not enough time for

the radon to reach in the samples (a time period of at least 30 days is needed). Therefore the measured concentrations of ^{226}Ra were lower by about 20%. Gamma spectrometric measurements were performed with two high resolution HPGe gamma spectrometers.The first one, the GMX type, made by ORTEC with nominal efficiency exceeding 36% and reslution less than 1.9 keV, was placed in a special low background schielding chamber with lead walls 12 cm thick. The second CANBERRA HPGe detector with nominal efficiency 22%, was placed in another schielding chamber with iron walls 25 cm thick. The spectra were led through the preamplifirier-amplifier chain to CANBERRA Ser. 35+ multi channel analyzer with two analog-to-digital converters, and with a memory containing 8192 channels. A modified version of the SAMPO program was used to process the spectra, in such a way that, besides the identified gamma-lines, it always presented spectral intensities of 20 selected isotopes. The samples were measured in cylindrical geometry, placed in sample containers with 67 mm diametar and 62 mm height.

In order to measure the radon emanation from soil, a metallic cylinder with Φ=13 cm and height H=70 cm with the upper part closed was digged in the soil. The top of the cylinder was at the soil surface level thus the charcoal canister with Φ=10.5 cm placed in the cylinder was exposed 65 cm below the surface. The open end of the canister was oriented upwards. The exposure time was 2 days. The indoor radon samples (n = 175) were collected on from various locations in Novi Sad in the period 1998 – 2003 year. The selection of locations represents the structure of living rooms in the area of Novi Sad. The measurements of radon activity concentrations were performed in summer and winter weather conditions.

The method of the adsorption on activated charcoal canister was applied [6] in some measurements of radon activity concentrations. The canister gamma activity (the activity of ^{214}Bi and ^{214}Pb radon daughters) was measured by means of high resolution germanium and NaI(Tl) scintillation spectrometers. The efficiency of the detectors was determined using the EPA ^{226}Ra Reference source. The canisters were exposed for two days. The typical time between the end of the exposion and the beginning of the measurement was about two hours. In order to achieve 5% statistical accuracy at 100 Bq/m^3 the time of measurement was usually 1 hour. In our measuring chamber the radon levels are very low (less than 5Bq/m^3) so radon fluctuations could not affect the results of most measurements.

The latest measurements, in spring 2003, were made by CR-39 track detectors on about 100 locations in Novi Sad flats and houses with exposure time of 90 days. CR-39, which is a clear 1 cm^2 stable plastic sensitive to the tracks of alpha particles, is the most widely used and accurate detector for radon measurements. The etching, evaluating and counting processes were performed in Hungary by Radosys Company.

Results

In Table 1. the results of gamma spectrometric measurements of buildings materials are presented together with the Building Material Index according the formula (1).

Tab. 1 Average activity concentrations of radionuclides in building materials

numbers of samples	building material	^{238}U	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	Build.Mat. Index
6	brick	54±23	40±16	43±19	526±149	1.7±1.4	0.51±0.18
1	siporeks	19±11	<2.7	9.9±1.0	196±15	1.4±0.5	0.106±0.009
1	gutter block	79±29	41.5±2.0	47±3	630±50	2.1±0.7	0.575±0.021
3	ceramic tiles	139±44	91±20	70±5	710±20	<0.8	0.93±0.12
1	glue for the ceramic tiles	22±21	21.8±1.5	9.9±1.1	178±15	<0.5	0.202±0.010
1	mass for the tiles polishing	11±5	1.6±0.7	8.1±0.6	79±8	<0.22	0.062±0.005
1	chalk	<9	<0.6	0.32±0.25	10.4±2.7	0.5±0.3	<0.003
2	cement	65±8	57±10	19.5±0.7	200±9	<0.8	0.40±0.04
1	parquet	<14	<4	3.2±1.5	44±12	<1.6	<0.019
2	granite	45±10	23±20	35±33	705±502	<0.4	0.5±0.4
2	tile	59.5±0.7	43.0±0.4	56±6	696±64	<0.4	0.63±0.04

In Table 2. some typical results of measurements on soil samples performed during 2002 year are presented. For different given locations in Novi Sad area, average activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were determined from the values on several micro locations.

Tab. 2 Average activity concentrations of radionuclides in soil samples from the listed locations for 2002 year

		ACTIVITY CONCENTRATIONS [Bq/kg]				
number of microlocations	locations in Novi Sad area	^{238}U	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
4	ŠANGAJ	47±4	32±3	39±7	501±71	9±7
4	PETROVARADIN	85±21	44±18	37±6	500±82	4±3
4	BISTRICA	39±2	33±4	37±5	538±94	4.9±2.9
4	TELEP	37±4	31±7	36±6	525±53	9±5
4	KLISA	54±10	34±6	38±10	514±136	3.9±1.5
4	ADICE	42±10	34±3	38±4	555±48	7.6±0.5
6	LIMAN	42±5	24±6	25±6	423±64	6±5

On Fig.1 results of measurements of emanated radon concentrations from soil in Novi Sad area are presented. On the base of this results (about 100 measurements) average, minimal and maximal emanated radon activities are calculated (Table 3).

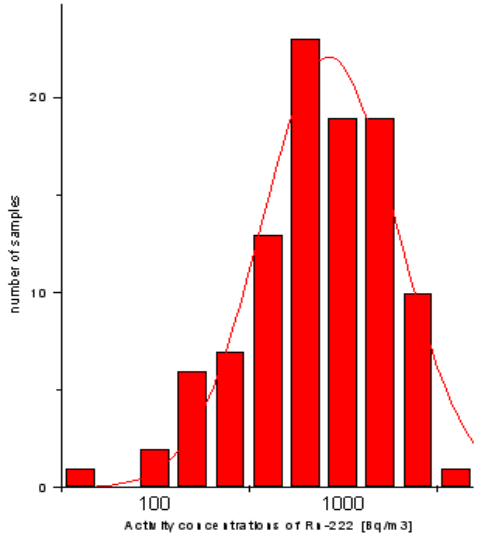

Fig.1 Frequency distribution of emanated radon activity concentrations during the period 1997-2003

Tab.3 Maximum (A_{max}), minimum (A_{min}) and average emanated radon activity concentration from soil A_{av} with standard deviation σA in Novi Sad area

radionuclide	A_{max} [Bq/m^3]	A_{min} [Bq/m^3]	A_{av} [Bq/m^3]	σA [Bq/m^3]
^{222}Rn	3247	44	989	719

On Fig.2 results of measurements of indoor radon concentrations in Novi Sad flats by charcoal cannisters are presented. The results of the statistical analysis of about 180 measurements are listed in Table 4.

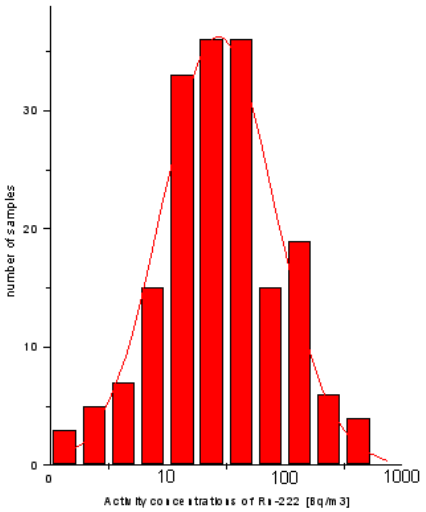

Fig.2 Frequency distribution of radon activity concentrations in Novi Sad flats during period 1992-2003

Tab.4 Maximum (A_{max}), minimum (A_{min}) and average indoor radon activity concentration A_{av} with standard deviation σA in Novi Sad flats

radionuclide	A_{max} [Bq/m^3]	A_{min} [Bq/m^3]	A_{av} [Bq/m^3]	σA [Bq/m^3]
^{222}Rn	391	2	50	69

The results of the latest measurements of indoor radon concentrations by CR-39 performed in spring 2003 (in about 100 flats and houses) are presented on Figure 3 and in Table 5.

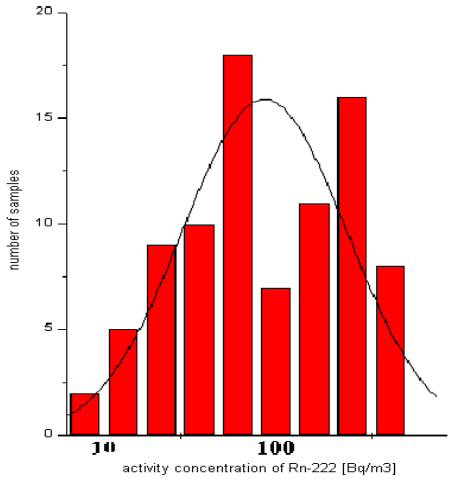

Fig 3. Distribution of indoor radon activity concentrations measured by CR-39 during the period December 2002 – March 2003

Table 5. Maximum (A_{max}), minimum (A_{min}) and average indoor radon activity concentration A_{av} with standard deviation σA in Novi Sad flats measured by CR-39 during period December 2002 – March 2003

A_{av} [Bq/m^3]	σ [Bq/m^3]	n number of samples	Amin [Bq/m^3]	Amax [Bq/m^3]
133	115	86	10	445

Discussion

In building materials (Tab.1), the activity concentration of the natural radionuclide ^{40}K was dominant. The only anthropogenic radionuclide in samples was ^{137}Cs. On the base of our results the building materials with the greatest radioactivity content are the ceramic tiles.

In soil samples (Tab.2), activity concentration of natural radioactive series of ^{238}U is in the range 20 – 70 Bq/kg which is the normal range for Panonian soil. The soil radioactivity results show that ^{238}U and ^{226}Ra activities are in equilibrium and there is no indication of depleted uranium presence [7]. The activity concentrations of ^{232}Th (in the range 17.8 - 52.6 Bq/kg) and the natural radionuclide ^{40}K are also at the normal environmental levels.The radionuclide ^{137}Cs, which originates from the accident of the nuclear power plant Lenin in Chernobyl in 1986, was identified in all soil samples and due to 30 years half-life it will be present for a long time in ecosystem of Eastern Europe.

The mean-value of radon activity concentration emanated from soil (Tab.3) is 987 Bq/m^3 with the standard deviation of 733 Bq/m^3. We emphasize that the results from nearby locations are significantly scattered, thus the radon emanation is very no uniform. Further no correlation was found between the ^{238}U – ^{226}Ra concentration and the radon emanation intensity. This means that the radon emanation from soil is mainly determined by deep soil cracks, soil porosity and underground water streams.

The mean value of indoor radon activity concentration (Tab.4) obtained by charcoal canisters is (50 ± 69) Bq/m^3. Only 5 % of the results exceed the 200 Bq/m^3 value accepted as an intervention level in Serbia and Montenegro. The highest results are obtained for flats on ground level. The owners of this flats and houses were advised how to solve the radon build-up problem. The lognormal distribution obtained proves the random nature of the radon build-up in the flats investigated.

The target locations of the newest investigations of indoor radon concentrations by CR-39 track detectors were old farmer houses in the suburbs region. The results of this measurement (Tab.5) yield the mean value of 133 Bq/m^3 with the standard deviation of 115 Bq/m^3. Almost 30% of the measurements are over the 200 Bq/m^3 and 1% of the measurements are significantly elevated indoor radon concentrations. The results are above the expectations for the low-land districts and show that dominant sampling in new city flats can seriously underestimate the radon buildup problem.

Conclusions

The results obtained confirm that radioecological problems at home building are not neglibile even in flat agricultural region. With detailed measurements and carefull selection satisfactory building materials can be found, just slightly contributing to the radon build-up problem. However bad ground isolation in the houses will cause significant radon build-up even on soils with moderate uranium content.

Acknowledgment

The authors acknowledge the financial support of the Ministry of Science, Technology and Developing of Serbia, in the frame of the project Nuclear Spectroscopy and Rare Processes (No 1859) and the City Development Institute of Novi Sad.

References

[1] О.С.Андреева, В.И.Бадьин, А.Н.Корнилов, *Природный и обогащенный уран,* Москва, Атомиздат, 1979.

[2] Таблицы физических величин, *Справочник под редакцией академика И.К.Кикоина,* Москва, Атомиздат, 1976.

[3] United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR). Ionizing radiation: Sources and biological effects. Exposures to radon and thoron and their decay products. 1982. Report to General Assembly, with annexes. 141-210

[4] EC, 1997: Radiation Protection 88. Recommendations for implementation of Title VII of the European Basic Safety Standards concerning significant increase in exposure due to natural radiation sources. European Commission. Office for Official Publications of the European Commission. Radiation Protection Series.

[5] Radon Legislation and National Guidelines. SSI report. Swedish Radiation Protection Institute. No 99: July 1999. ISSN 0282-4434

[6] U.S. Environmental Protection Agency. National Residential Radon Survey: Summary Report. EPA-402-R-92-004.1992

[7] I.Bikit, J. Slivka, D. Mrdja, N. Zikic-Todorovic, S. Curcic, E. Varga, M. Veskovic, Lj. Conkic: Simple Method for Depleted Uranium Determination, to be published in Japanese Journal of Applied Physics (JJAP), Tokyo, Japan