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RADON INDOOR CONCENTRATIONS AND ACTIVITY OF RADIONUCLIDES IN BUILDING MATERIALS IN SERBIA

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Abstract. *The paper presents the results of radon indoor concentrations measurements in homes and public institutions (kindergardens) in Belgrade, as well as the results of the radionuclides content determination in building materials from Serbia, during the eighties and up to the mid-nineties of the 20th century. The activity of the radionuclides was determined on an HPGe detector (ORTEC, relative efficiency 20%) by standard gamma spectrometry. Radon indoor concentrations on the grab samples of air were determined by alpha scintillation technique (Lucas scintilation cell with ZnS/Ag). Mean effective dose equivevalents of radon and its progenies in closed space were estimated at 0.074 mSv for radon and 1.22 mSv for its short-lived daughters, in total 1.3 mSv.*

Key words: *radon, radioactivity, building materials, gamma spectrometry*

INTRODUCTION

Radiation exposure due to natural radionuclides in building materials, as well as radon concentrations in closed space, was recognized as a significant cancer risk for the general population only in the early seventies in the 20th century. In Europe the annual effective dose equivalent from all sources of radiation in the environment is estimated to 3.3 mSv and doses of natural radiation account for about 80% of this value, while the average annual exposure to indoor radon, thoron and their short-lived daughters are estimated to 1.6 mSv. The most significant natural radionuclides in building materials are ^{40}K, and ^{238}U, ^{235}U, ^{232}Th and their progenies. Average concentrations of thorium and uranium in soils are 4-12 g/t, while for comparision, the average concentrations of rare metals are within the range of 0.02-0.05 g/t [1,2,3].

The content of natural radionuclides in building materials is caused by many factors: geological origin and composition of soil, its density and porosity, content of water in

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soil, diffusion rate and permeability rate, rate of emanation and exhalation, etc. Thus, while blocks of fired clays have insignificant percentage of radioactive elements, cement contains up to 5.9 g/t of uranium, 20g/t of thorium and $2x10^4$ g/t of potassium-40. Therefore, there are significant differencies in the effective dose equivavalent in closed space due to the type of building material and construction of the building object: it is within the range of nearly 0 mSv/year for objects made of wood, up to 0.2 mSv/year for objects made of concrete, 0.8 mSv/year for stone objects and objects containing phosfogypsum and 1.7 mSv/year for objects made of slag panels [4,5].

In some of the building materials the content of radionulcides is higher than in soils, for example in granite, slate or vulcano rocks. The higher content of radionuclides could be due to the use of secondary raw materials used in construction, like electrofiltering ash from coal burning processes in thermo-electric power plants, etc. Phosphogypsum and other materials expressing the so-called *technologically enchaned radiactivity* could increase radon concentrations in closed space for factor of 100 and more. Man-made radionuclides, mainly ^{137}Cs, could be found in building materials as a result of fallout deposition on soil or sand surfaces [5].

Radon is a radioactive noble gas, colourless and odourless, chemicaly inert, but highly dissolable in water and non-polar solvents, with the half-life of 3.84 days. All its progenies are short-lived: 0.2 ms – 26.8 min, and among them, the most important are alpha emitters ^{218}Po, ^{214}Po and ^{210}Po. They attach to aerosols, thus entering the lungs where alpha particles damage the basal cells of the lung tissue. Migrating from soil and underground waters, radon enters the atmosphere and concentrate in closed space. Main radon entries in closed space (homes and public buildings) are cracks in walls and floors, water and ventilation pipes and emanation from building materials [1].

Mean radon concentrations in closed space are 2-15 times higher in winters than in summers. Due to the type of building materials they are in the range of 20 Bq/m^3 in objects made of siporex/light concrete and natural bricks, and up to 60 Bq/m^3 in objects made of slag concrete blocks. In Serbia, the highest radon concentrations were measured in air in Niška Banja (140-1,100 Bq/m^3, even above 10,000 Bq/m^3 in some of the spa objects) [6,7].

First measurements of radon in the former Yugoslavia were performed in uranium mines of Žirovski vrh, and in the spas and karst caves in Slovenia in the early eighties of the 20th century [6]. Also, during the eighties and up to the mid nineties radon indoor concentrations were measured in homes and public buildings in Serbia, but not sistematically. Therefore, there was no sufficient information to form a database for *radon mapping.*

Worldwide, sistematic measurements of radioactivity in building materials and estimation of exposures from radon in closed space – homes, flats and offices – started in the early eighties in the last century, while risk of radon, thoron and their short-lived progenies was recognized as a global problem and a significant cause of cancer increase in general population in the late seventies. Starting in the seventies, there were numerous studies confirming indoor radon as a cause of over 20,000 lethal lung cancer cases in EU every year, making about 9% of all lethal lung cancer cases per year in EU and about 2% of lethal cancer cases per year in EU in general. The majority of European countries, as well as Canada and USA, proclaimed intervention levels for exposures from radon in closed space and concentrations of radionuclides in building materials. These issues were also regulated by numerous publications and recommendations by international organizations: World Health Organization (WHO, 1985), International Commission For Radiation Protection (ICRP, 1985), United Nation Commission For Atomic Radiation Research (UNCEAR, 1988), International Atomic Energy Agency (IAEA, 1984), etc. According to these recommendations the general population should not be exposed to more than 0.7 mSv of radiation from building materials, therefore the total gamma radiation index for radionuclides ^{40}K, ^{226}Ra and ^{232}Th should not be higher than 1. Most of the European countries accepted the recommendations, although some of the Scandinavian countries facing specific climate conditions and geological soil characteristics allowed higher radon indoor concentrations (up to 200 Bq/m^3 for future buildings, the level accepted (mz) by majority of the European countries, intervention levels up to 800 Bq/m^3 for built and inhabited buildings, due to purpose). To achieve a common approach to the radon issue on the global level, International Radon Committee (IRC) and Radon Research Centers (RRC) Network were established during the eighties in the 20th century [8,9].

Although soil is considered as a main source of radon in closed space, some building materials and construction of building could significantly contribute to a higher level of natural radiation indoors. That is specially the case in European countries, while in USA and Canada soil and its geological characteristics contribute mainly to radon exposure in closed space.

In our country, up to the mid-nineties of the last century there were no standards or metrological instructions/procedures directly defining this issue, but it was indirectly and partly defined in legal regulations considering protection of ionizing radiation and environmental protection (Law on Environmental Protection, 1991; Law on Ionizing Radiation and Nuclear Safety, 1991; Law on Air Contamination, 1973,1977,1989; Regulations on Maximum Levels of Radioactive contamination of the Environment and Decontamination, 1987; Regulations on Maximal Permissable Concentrations of Contaminants in Air, 1978; Law of Protection Against Ionizing Radiation, 1996). Also, basic criteria for setting up and accreditation of laboratories involved in testing and control of radioactivity in building materails were regulated by the Law on Standardization in 1988 and 1991, and by the international standards *EN45001, EN45002* and *EN45011* (CEC, Luksemburg, 1989). These criteria made a framework for organization rules concerning laboratory spacing and staff, measuring procedures and instrumentation characteristics, calibration procedures, sampling, quality control and quality assurance. Still, there was no precisely regulated and defined monitoring program for radon survey in open and closed space and procedure to estimate dose equivalents due to inhalation of radon, thoron and its progenies [10].

EXPERIMENT

Radon concentrations were measured in Belgrade during the fall of 1990 and in the spring of 1991. Radon indoors concentrations were measured by alpha scintillation technique on the *grab* samples of air, between 05.00 a.m and 08.00 a.m, in rooms that were closed during the night for at least 12h before sampling. The scintilation cell was filled with air at sampling sites and transported to Laboratory of Radiation Hygiene at the Faculty of Veterinary Medicine in Belgrade, where alpha activity was measured after 3 h on IGMA Rn1 counter (IJŠ, Lubljana). The alpha scintillation cell (Cu cillinder 1.5 dm^3, coated with ZnS/Ag) was made in the Institute "Jožef Štefan", Ljubljana, and calibrated with standard radium solution and reference scintillation cell according to the standard procedure [11]. Cell background was 0.5-1.5 min^{-1}, and cell constant was in the range of 0.0019-0.0022 Bq^{-1}s^{-1}m^3. Counting interval was 30-60 minutes, and lower limit of detection (LLD) was 10-30 Bq/m^3.

Radon progenies were measured by the portable alpha spectrometer EDA-300, with air being continually pumped through the filter, and alpha particles counted every 30 minutes. In some cases, *alpha track* detectors have been exposed for 3 months and radon average indoor concentrations were determined. Gama dose rate was measured by a portable dose rate-meter (Gama Dose Rate Meter, ASP-1 Eberline).

Samples of raw building materials and final products (siporex light concrete, sand, gravel, cement, quartz sand, lime) were sampled at the production site, in the vicinity of Belgrade, homogenized and dried to constant mass at 105°C. The density of the samples was 0.3-1.3 g/cm^3, and the masses were in the range of 200-800 g. Samples were stored in standard Marinelli beakers (volume 0.5 l), the beakers were sealed with wax and left for 4 weeks to reach the radioactivity equilibrium.

Samples of clay originated from the open pit Košarno (geological origin tertier), 15 km from Mladenovac, in the mountain Bukulja. In the geological profile of the pit one could recognize 4 main types of kaolin clay G1, G2, G3 and G4, a surface fire clay layer CG and a layer of green clay ZG. The mineral content of the clay was determnined by X ray diffraction analysis, and the chemical content by standard differential termographimetry. For activity measurements clay was prepared according to standard gamma spectrometry sample preparation procedure.

The activity of the radionuclides was determined on an HPGe detector (ORTEC, relative efficiency 20%), by standard gamma spectrometry. The activity of radium and thorium was determined by their daughters ^{214}Bi, ^{214}Pb and ^{228}Ac, respectively. The activity of ^{235}U was determined on 185,7 keV, corrected for ^{226}Ra on 186 keV. Geometric efficiency was determined by a secondary reference soil standard (National Office of Measures NOM, Budapest) spiked with ^{22}Na, ^{57,60}Co, ^{88}Y, ^{133}Ba and ^{137}Cs (activities 122-355 Bq, measuring uncertainty 3%). Counting interval was 200.000 - 400.000s, total error of the method up to 15%.

RESULTS AND DISCUSSION

Radon in homes and kindergartens in Belgrade in 1990/1991

The results of radon indoor concentrations (Bq/m^3) and gamma dose rate (nGy/h) measurements in 167 dwellings (family houses, multistoried houses) in Belgrade are presented in Table 1. Minimal (Min) and maximal radon indoor concentrations (Max), arithmetic means (Mean), standard deviation (SD), all in Bq/m^3 and cofficient of variation CV (%) are presented. The values of gamma dose rate in air (nGy/h) are presented, too.

Fig. 1 presents distribution of the mean radon indoor concentrations measured in Belgrade dwellings during 1990/1991 for different sampling sites around the city (munnicipalities: Stari Grad 1, Čukarica 2, Vračar 3, Novi Beograd 4, Savski Venac 5, Zemun 6, Voždovac 7, Palilula 8, Rakovica 9, Zvezdara 10).

Table 1. Radon indoor concentrations and gamma dose rate in Belgrade dwellings

Radon concentrations (Bq/m^3)					Gamma dose rate (nGy/h)				
Min	Max	Mean	SD	CV(%)	Min	Max	Mean	SD	CV(%)
11	218	58	41	69	72	176	108	23	21

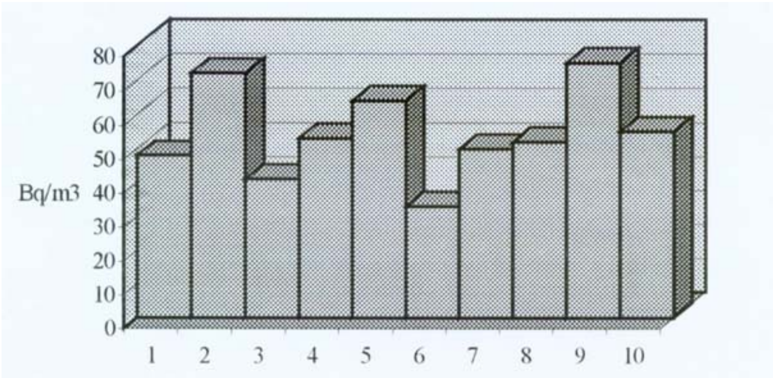

Fig. 1. Distribution of mean radon indoor concentrations in Belgrade:
(Numbers 1-10 mark the different municipalities in Belgrade)

Mean radon indoor concentrations in Belgrade dwellings were generally under 200 Bq/m^3, the value adopted as an action level for most of the European countries, while concentrations above this value were measured only in less than 5% of the surveyed buildings. There were significant differencies in radon concentrations between the buildings (69%), as expected, considering building materials, age of the building, geological composition of soil and site configuration. Maximal concentrations were measured in the buildings on the littered sliding soils with intense circulations of underground gases, while minimal concentrations were found on the sites with higher percentage of clay and water.

Mean radon concentration (58 Bq/m^3) was introduced into the Jacobi-Eisfeld model to estimate average effective yearly dose equivalent for radon and its short-lived progenies. Model parameters were the following: solubility factor for soft tissue 0.4, alpha radiation quality factor 20, weight factors for lung 0.12 and 0.88 for soft tissue, mean probable residence time 0.8 (equivavalent of 7000 h per year in closed space), and mean yearly exposure to short lived radon daughters 0.24 WLM. This gave an estimated value of 0.074 mSv for the effective yearly dose equivalent for indoor radon and 1.22 mSv for radon short lived daughters, in total 1.3 mSv [12].

Distribution of radon indoor concentrations due to the type of building material is presented in Table 2. Building materials correlated with age and type of the building: brick houses were mainly detached family houses built before the II World War (1), buildings made of bricks and concrete blocks were multistoried flat houses built between the fifties and seventies in the last century (2), buildings made of siporex light concrete were multistoried flat houses build during the eighties (3). Minimal radon concentrations were measured in siporex light concrete buildings, as expected considering the concentrations of natural radionuclides in siporex concrete (Table 4 and Fig.2b).

Table 2. presents the number of objects (%) made of individual building materials and measured radon indoor concentrations due to the building material. Table 3. presents vertical distribution of radon indoor concentrations (as no significant differencies between radon indoor concentrations on second and higher flooors were found, average values for second and higher floors are presented in the table).

Table 2. Radon indoor concentrations (Bq/m$_3$) in houses of different building materials

Material	Objects %	Min	Max	Mean	SD	CV (%)
Brick	46	11	192	54	44	81
Brick/concrete	34	12	218	53	44	83
Siporex	12	13	82	43	18	42
Mixed	8	12	168	48	28	58

Differences in radon indoor concentrations measured in buildings on the same site were under 10%, while in multistoried houses built in the eighties the differencies were only 2-3%, indicating building material to be the main source of radon in Belgrade dwellings. Average radon indoor concentrations measured in Belgrade dwellings were significantly lower than the intervention levels in USA (150 Bq/m^3), EU and Australia (200 Bq/m^3), and especially in Canada and Scandinavian countries (800 Bq/m^3) [12].

Table 3. Vertical distribution of radon indoor concentrations (Bq/m^3)

Floor	% objects	Min	Max	Mean	SD	CV(%)
Groundfloor	53	11	218	64	47	73
I floor	33	11	196	40	31	78
II & higher	14	20	38	29	6	21

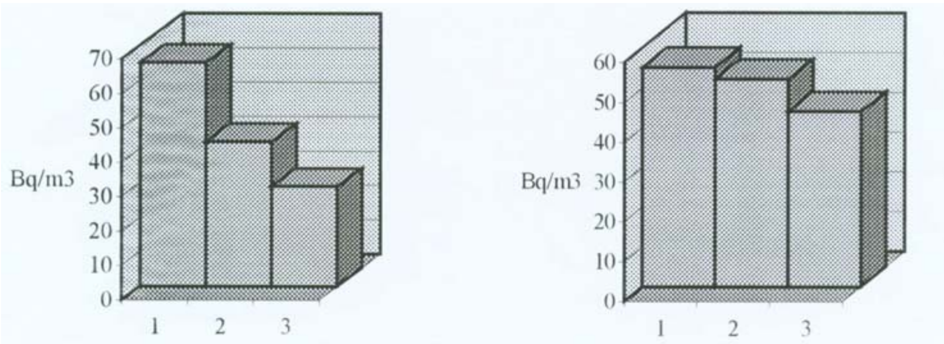

Fig. 2. Radon indoor distribution due to floor (a) and age of the building (b)

Distribution of radon indoor concentrations in Belgrade dwellings due to floor and age of the building is presented on fig. 2a and fig. 2b, respectively. (Fig. 2a: 1 – ground floor, 2- I floor, 3 – II and higher floors. Fig. 2b: 1 – houses built before 50ties, 2 – houses built from 50ties up to 70ties, 3 – houses built during the 80ties of the 20th century).

The vertical distribution of radon indoor concentrations presented in Table 3. pointed to significant differencies in average radon concentrations measured in ground floor and higher floors, but also between values measured on the same floor in different buildings, especially for the ground floor and the I floor. This is probably due to the differencies in the geological characteristics of soil, but also to the construction of the building. On average, radon concentrations were decreasing from the ground floor towards higher floors for about 20% per floor.

During fall of 1990 and spring of 1991 radon concentrations were measured in kindergartens in major cities in former Yugoslavia [13]. The results are presented in Table 4.

Table 4. Radon indoor concentrations (Bq/m^3) in kindergartens in former Yugoslavia

Town (Numbers of kindergartens / children)	Min	Max	Mean
Ljubljana (130 / 16650)	10	5606	177
Maribor (97/ 7200)	11	945	107
Nova Gorica (39/ 2250)	9	267	44
Kranj (17/ 1650)	8	144	38
Osijek (21/ 2900)	10	178	67
Sarajevo (41/ 5350)	11	148	27
Belgrade (104/ 15000)	12	227	59

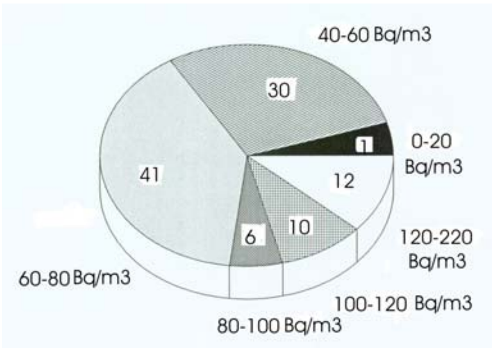

Fig.3. Radon indoor concentrations in kindergartens in Belgrade
(the numbers on pie mark the numbers of kindergartens)

As presented in Table 4. the highest mean radon concentrations were measured in kindergartens in Ljubljana and Maribor, where the maximal radon concentrations have been recorded, too. That is due to the building materials (materials containing higher amounts of natural radionuclides), construction (older buildings with cracks in floors), position (basements or ground floors) and soil characteristics (cinder, sedimentary formations) [13].

As for kindergartens in Belgrade, the variations between radon indoor concentrations in different kindergartens were about 66%, but only in 12 of the buildings radon concentrations above 100 Bq/m3 were found. Ranges of indoor radon concentrations in Belgrade kindergartens and the number of objects, are presented in Fig. 3.

Simultaneously, in Maribor, Osijek and Belgrade in kindergartens where radon indoor concentrations exceeded 150-200 Bq/m3, alpha track detectors were exposed for 3-4 months [13]. Mean radon indoor concentrations measured by alpha track detectors were 1.5 to 2.0 times lower than instantaneous radon indoor concentrations measured by alpha scintillation technique. This, considering the mean radon indoor concentrations for radon mapping one should consider the methodology, too.

Natural radionuclides in building materials from Serbia

Contents of natural radionuclides ^{226}Ra, ^{232}Th, ^{40}K and ^{235}U in raw building materials and final products – siporex extended/light concrete produced in Serbia in the late 80ties and up to mid-90ties of the 20th century are presented in Table 5. The results indicate that on the average the level of natural radionuclides in building materials from Serbia were lower than in similar building materials from the region and therefore, lower radon indoor concentrations due to excalation from walls should be expected [7].

Table 5. Activity of natural radionuclides (Bq/kg) in building materials

Material	^{226}Ra	^{232}Th	^{235}U	^{40}K
Siporex concrete	9.4 ± 0.9	5.4 ± 0.6	121 ± 10	0.70 ± 0.08
Cement	49 ± 4	17 ± 2	238 ± 19	3.6 ± 0.3
Quartz sand	4.7 ± 0.5	3.5 ± 0.4	84 ± 6	0.5 ± 0.04
Sand/gravel	12 ± 1.0	12.1 ± 1.0	241 ± 20	1.0 ± 0.1
Unhydrated lime	4.3 ± 0.5	1.0 ± 0.3	43 ± 4	0.45 ± 0.40

As presented in literature [5,7] the activity of natural radionuclides in building materials produced in Serbia during the eighties of the 20th century was in the range of 44-360 Bq/kg for ^{226}Ra (max values measured in slag blocks) and 70-530 Bq/kg for ^{40}K (max values measured in bricks). Activity of ^{137}Cs deposited on the surface during the Chernobyl nuclear plant acident, in April 1986, was in the range of 4–155 Bq/kg (max values measured in bricks).

The mean activities of natural radionuclides measured in building materails produced in EU and former Yugoslavia in the last decades of the 20th century did not differ significantly from the values measured in building materials produced in Serbia. Maximal concentrations of ^{226} Ra and ^{232}Th were measured in electro-filtering ashes and phosphogypsum from Slovenia (1900 Bq/kg ^{226}Ra) and some building materials from Germany, while minimal concentrations of natural radionuclides were detected in building materials produced in Scandinavia, mainly in siporex light concrete and natural bricks. Maximal concentrations of ^{40}K were measured in different slate containing materials – up to 900 Bq/kg [7,14].

Radionuclides in clays used in ceramic industry in Serbia

The results of natural radionuclides determination in clays used in ceramics industry in Serbia (Kosarno open pit) up to the mid-nineties of the 20th century, are presented in Table 6. (the values presented in the table are the means from multiple measurements, standard deviation being within the total error of the method – up to 15%).

Table 6. Radionuclides (Bq/kg) in clays used for ceramics in Serbia, 1990/1995

Clay layer	^{226}Ra	^{232}Th	^{40}K
CG	34	63	561
G1	174	73	485
G2	64	66	519
G3	82	82	875
G4	59	70	813
ZG	61	60	783

CG - surface layer of brick clay, G1,G2,G3 and G4 – kaolin clays, ZG – green clay

To compare the contents of radionuclides in clays originating from Serbia and the one in clays from the region and elsewhere, we have calculated the equivalent indices for radium Ra(eq), thorium Th(eq) and potassium K(eq), as well as the OECD index. The indices have been calculated according to equations [7]:

$$\begin{aligned} Ra\,(eq) &= A(Ra) + 1.43\,A(Th) + 0.077\,A(K) \\ Th\,(eq) &= A(Th) + 0.70\,A(Ra) + 0.054\,A(K) \\ K\,(eq) &= A(K) + 18.46\,A(Th) + 13.24\,A(Ra) \end{aligned} \tag{1}$$

where A is the activity of the radionuclide (Bq/kg).

The calculation is based on the assumption that 0.37 Bq/g of ^{226}Ra, 0.26 Bq/g of ^{232}Th and 4.8 Bq/g of ^{40}K effectuate equal gamma dose rate equivalent.

OECD index was calculated according to[7]:

$$\text{OECD index} = [A(K)/1500] + [A(Ra)/150] + [A(Th)/100] \tag{2}$$

The results of calculations are presented in Table 7.

The calculated values of the equivalent indices for natural radionuclides and OECD index for clays originated from Kosarno open pit were on the upper limit of the average values for clays used in ceramic industry in most of the European countries and Middle East. This was due to higher values of natural radionuclides found in Kosarno clays, mainly for ^{40}K. The contents of natural radionuclides in clays used at the end of the 20th century in the ceramic industry in EU and Middle East were in the range: 20-60 Bq/kg for ^{226}Ra, 20-90 Bq/kg for ^{232}Th and 340-1000 Bq/kg for ^{40}K [15,16].

Table 7. Equivalent indices and OECD index for Kosarno clays

Type of clay	Ra (eq)	Th (eq)	K (eq)	OECD index
CG	167	117	2294	1.23
G1	288	202	3983	2.03
G2	202	139	2587	1.45
G3	251	176	3278	1.82
G4	222	155	2688	1.63
ZG	207	145	2687	1.53

CONCLUSION

The study was a part of the research that should present the base for the *Program of Quality Control and Radioactivity Monitoring in Building Materials,* as well as for the *National Radon Program.* The program should have comprised standardization of sampling procedures, measuring techniques and calibration procedures to measure activity of natural radionuclides in building materials and to form a data base for estimating exposure from radon in closed space. Eventualy, intervention levels for radon for built objects and those under construction should have been established and radon mapping have been performed.

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MERENJA KONCENTRACIJE RADONA U ZATVORENOM PROSTORU I AKTIVNOSTI RADIONUKLIDA U GRAĐEVINSKIM MATERIJALIMA NA TERITORIJI REPUBLIKE SRBIJE

Dragana Popović, Dragana Todorović

U radu se daju rezultati odredjivanja sadržaja radona u vazduhu u zatvorenom prostoru u stambenim i javnim objektima (dečijim vrtićima) u Beogradu, kao i rezultati odredjivanja sadržaja radionuklida u gradjevinskim materijalima sa teritorije Srbije, tokom osamdesetih i do sredine devedesetih godina 20.veka. Aktivnost radionuklida odredjivana je na HPGe detektoru (Ortec, relativna efikasnost 20%) standardnom metodom spektrometrije gama zračenja. Koncentracije radona u vazduhu odredjivene su alfa scintilacionom tehnikom, pomoću Lukasove scintilacione ćelije sa ZnS/Ag (IJŠ, Ljubljana). Na osnovu mernih podataka procenjena je srednja efektivna ekvivalenta doza za radon i njegove kratkoživeće potomke u zatvorenom prostoru: 0.074 mSv i 1.22 mSv, respektivno, odnosno ukupna doza od 1.3 mSv.