HIGH ANNUAL RADON CONCENTRATION IN DWELLINGS AND NATURAL RADIOACTIVITY CONTENT IN NEARBY SOIL IN SOME RURAL AREAS OF KOSOVO AND METOHIJA

by

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Scientific paper
DOI: 10.2298/NTRP1301060G

Some previous studies on radon concentration in dwellings of some areas of Kosovo and Metohija have revealed a high average radon concentration, even though the detectors were exposed for three months only. In order to better design a larger study in this region, the annual measurements in 25 houses were carried out as a pilot study. For each house, CR-39-based passive devices were exposed in two rooms for the two consecutive six-month periods to account for seasonal variations of radon concentration. Furthermore, in order to correlate the indoor radon with radium in nearby soil and to improve the knowledge of the natural radioactivity in the region, soil samples near each house were collected and ^{226}Ra, ^{232}Th, ^{40}K activity concentration were measured. The indoor radon concentration resulted quite high from the average (163 Bq/m^3) and generally it did not differ considerably between the two rooms and the two six-month periods. The natural radionuclides in soil resulted to be distributed quite uniformly. Moreover, the correlation between the ^{226}Ra content in soil and radon concentration in dwellings resulted to be low (R^2 = 0.26). The annual effective dose from radon and its short-lived progeny (5.5 mSv, in average) was calculated by using the last ICRP dose conversion factors. In comparison, the contribution to the annual effective dose of outdoor gamma exposure from natural radionuclides in soil is nearly negligible (66 Sv). In conclusion, the observed high radon levels are only partially correlated with radium in soil; moreover, a good estimate of the annual average of radon concentration can be obtained from a six-month measurement with a proper choice of exposure period, which could be useful when designing large surveys.

Key words: radon, soil radioactivity, radiological impact

INTRODUCTION

Many studies on the indoor radon have been carried out in Balkan region [1]. Also, in the last decade, a considerable attention has been given to the indoor radon survey in Kosovo and Metohija, although it limited to some specific regions [2-7].

However, evaluations of radon concentration in these studies (as in other all around the world [8]) were generally based only on the three-month measurements, and annual averages could not be evaluated because adequate correction factors for the seasonal variations were not available. Therefore, in order to obtain an unbiased evaluation (*i. e.* not affected by seasonal variations) of annual radon concentration in dwellings of Kosovo and Metohija, a new measurement campaign in the area was necessary and the measurement of radon concentration had to be conducted for a total of 12 months. To this purpose, a pilot study has been conducted (from December 2009 to December 2010) in 25 houses distributed in 13 villages of 8 municipalities of the region. In particular, two "mixed" six-month exposure periods were selected, *i. e.* periods with overall similar mixture of cold and warm months, in order to test if each of these periods could be used to estimate the annual average with a low bias.

Moreover, an effort has been made in this study to evaluate the correlation between the radon level in

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dwellings and the ^{226}Ra concentration in soil samples taken close to the selected dwellings. This is interesting because whereas the soil is generally the main source of radon concentration in indoor air, the correlation between the indoor radon and radium in soil is often not very strong [9], although in some situations it resulted to be significant [10], and not many studies have been carried out trying to correlate the indoor radon and radium content in nearby soil (*e. g*., [11, 12]). Finally, in order to increase the knowledge on the natural radioactivity in the region (which could be useful also for comparing the soil contamination from artificial radionuclides), the other main natural radionuclides (*i. e.* ^{232}Th and ^{40}K) in soil were also measured.

In summary, in this paper methodology and results of the study are presented as regards: (a) the annual radon concentration and comparison of the two six-month periods, (b) the natural radionuclide content in the soil, including the correlation between radium in soil and indoor radon, and (c) the evaluation of the effective dose for the population living in these areas.

Study area and its geology

The studied area, Kosovo and Metohija, is located between latitudes 41° 51' and 43°15' N and longitudes 20° 01', and 21°48' E. The 25 houses involved in the study are distributed in 13 villages belonging to 8 different municipalities of the South-Eastern and Central Kosovo.

The terrain is plain, with hilly and mountainous landscapes of average height of over 500 m above the sea level. The continental climate indicates cold winters and warm summers, windy springs and rainy autumns. Temperature ranges from –10 °C to 30 °C.

The complex geology of studied area is due to the different periods of formation that range from Cambrian to Quaternary. It is rich in volcano-sedimentary formations and ophiolitic mélanges. The deposits fill numerous depressions with marl-sandy clays and sands bellow the Neocene and Quaternary overstep sequence with lignite deposits [13]. Formations set in magmatic and orogenic phases include few belts of ophiolites ranging in age from Jurassic to Cretaceous. These ophiolitic mélanges consist of marly limestones, conglomerates, sandstones, massive limestones, silts, carbonates and clastics. From Cambrian to Triassic became phyllites, calc-schists, marbles, dolomites, schists, and gneisses.

EXPERIMENTAL PROCEDURE

Indoor radon measurements

Aiming to cover the biggest possible field terrain regarding radon survey in Kosovo and Metohija, the new cycle of radon survey in the region (including new homes and locations) started by investigating 25 homes in the period between 2009 and 2011. In order to obtain the permission to measure radon concentration, the houses of some students from the University of Priština were selected for this survey. Although these houses are not randomly selected, they are still appropriate for the purposes of this study. Only in two locations the measurements were repeated, *i. e.* in Gracanica and Dobrotin, due to the previously measured high indoor radon concentrations [6, 7]. The houses are rural types houses with self-contained ground-floor with or without concrete slab and of average age of about 30 years. The solid fuel heating and natural ventilation are typical for these houses. In addition to the sandy soil or clay, somewhere even the stone is under and around the house's basement, mainly. The study area gravitates toward two active mines of lead and zinc [3].

The indoor radon measurements were carried out by exposing passive device, which consisted of a small dome-shaped diffusion chamber of 4.5 cm diameter and 2 cm height, made of conductive plastic, housing on the bottom a CR-39 nuclear track detector (2.5 cm   2.5 cm, 1 mm thick) model TASTRAK made by Track Analysis Systems Ltd., for the two consecutive six-month periods, from December 2009 to June 2010 and from June 2010 to December 2010. The devices were placed in the kitchen (or living rooms) and bedrooms, at a distance of about 30 cm from walls (in order to reduce the thoron contribution to the detector track density) and at the (a) height of 1.5 to 2.0 m above the floor. The first period relates to winter-spring, and the second relates to summer-autumn, so that the similar radon concentration values could result, on average, for the two periods. The tracks of alpha particles in the detectors were visible after chemical etching in a 6.25 N NaOH solution at 98 ºC for 1 h. The readout of alpha particle tracks was carried out by an automatic track analysis system (Politrack, Italy). The etching, track counting and radon concentration assessment were performed by the Italian National Institute of Health.

Measurement of radioactivity in soil

During the indoor radon measurements soil samples from undisturbed area near the houses were collected in 15 sampling sites. For neighboring houses, which were not but several meters far from each other, the soil samples were taken from one joint sampling site, *i. e.* of one square meter. Samples were taken by shovel from the three different depths. For the superficial samples, about 200 ml surface soils from each angle of a one square meter and from a center of this square were taken up to the 5 cm depth, whereas the other two one litre soil samples were taken both at the center of the one square meter at 10-20 cm and 20-40 cm depth. All samples were closed in plastic bags and transported to the laboratory. After removing the stones and roots, each sample was dried in an oven at temperature of 100-110 °C for about two hours to eliminate the moisture. Then the samples were crushed and sifted on sieve < 2 mm aiming at getting volume of 450 ml. The samples were stored in Marinelli beakers, closed by silicone sealant and left for more than 4 weeks to achieve secular equilibrium between radium and its decay products. The gamma spectrometric system used for the analysis of the samples consists of a coaxial high-purity germanium detector (GEM30-70, ORTEC) with relative efficiency of 32% at 1.33 MeV (^{60}Co) and of multichannel analyzer. The system had an energy resolution (FWHM) 1.69 keV at 1.33 MeV (^{60}Co) and 725 eV at 122 keV (^{57}Co). The detector was settled in a 10 cm thick lead shelter in order to reduce the background. Background and each soil sample were measured 10800 s under the same conditions.

The gamma-activity of radionuclides was determined through their intensity of emission lines in the spectrum after the background subtraction. Activity concentration of ^{226}Ra was determined as weighted average activity obtained from the two separate gamma photopeaks of its decay products, *i. e.* ^{214}Pb at 351.9 keV, and ^{214}Bi at 609.3 keV. The activity concentration of ^{232}Th was determined by the gamma photopeaks of ^{228}Ac (at the energies of 911.1 keV and 968.9 keV) and ^{208}Tl (at the energies of 583.0 keV and 860.6 keV). The photopeaks are created by gamma rays from de-excitation of decay products (gamma rays following β-decay of ^{214}Pb (to ^{214}Bi), ^{214}Bi (to ^{214}Po), ^{228}Ac (to ^{228}Th), ^{208}Tl (to ^{208}Pb)). From the photopeak at energy line of 1460.7 keV, gamma-activity concentration of 40 K was obtained.

RESULT AND DISCUSSION

Indoor radon

Out of the 25 monitored houses, full and validated measurements (*i. e.* both rooms and both periods) were available for 17 houses, whereas partial measurements were available for the remaining 8 houses: valid measurements for both rooms during one measurement period were only available for 4 houses, and measurements during both periods in one room were available for 4 houses. An anomalous radon concentration value found in one home, in one of the two rooms during the second period, has been excluded from this analysis, and the house was considered in the group of houses where the measurements during both periods were available only in one room.

The summary statistic of measurements carried out during the two exposure periods, in the two monitored rooms, is presented in tab. 1. Out of the 25 measured houses, the houses with both rooms measured were 22 and 18 for the first and second exposure period, respectively. For each period, the average indoor radon concentration in each house was estimated as the arithmetic mean of results in the two rooms.

The differences between the two measured rooms in each house were investigated. The ratios between radon concentration measured in the kitchen or living rooms and that measured in bedrooms follow the expected lognormal shape, with a geometric mean for the first and the second period of 0.9 and 1.0, respectively, and a geometric standard deviation of 1.7 and 1.6, respectively. Therefore, no significant systematic difference appears between radon concentrations in the two room types, as confirmed by the Kruskal-Wallis test.

According to the above result, in case of a missing measurement in a room, the available measurement in the other room was assumed to be representative for both ones. In this way it was possible to estimate the house average radon concentration, during the two six-month periods, for 25 and 21 houses respectively (see tab. 1).

The ratio between radon concentration measured in the two periods was analyzed in order to ver-

Table 1. Summary statistic of indoor radon concentrations measurements in the two periods of exposure and of the annual indoor radon concentrations

Period	Location	N	Indoor radon concentration [Bqm^{-3}]						
			Min	Max	Med	AM	SD	GM	GSD
First (Dec. 09-June 10)	Kitchen or living room	24	49	423	139	173	91	151	1.7
	Bedroom	23	27	380	136	161	94	136	1.9
	House average[a]	22	38	336	136	165	82	145	1.7
	House average[b]	25	38	336	138	169	79	150	1.7
Second (June 10-Dec. 10)	Kitchen or living room	20	44	359	133	173	97	147	1.8
	Bedroom	19	32	489	123	170	119	138	1.9
	House average[a]	18	46	424	157	179	101	153	1.8
	House average[b]	21	44	424	130	166	99	140	1.8
Complete year (Dec. 09-Dec. 10)	Kitchen or living room	20	47	383	161	179	94	155	1.8
	Bedroom	18	46	424	157	179	101	153	1.8
	House average[a]	17	61	378	164	179	88	159	1.7
	House average[c]	25	41	378	138	136	84	143	1.7

AM – arithmetic mean, SD – standard deviation, GM – geometric mean, GSD – geometric standard deviation (dimensionless)
[a] houses with valid measurements in both room, [b] houses with valid measurements in 1 room, [c] houses with valid measurements in 1 room and 1 period

ify if the two six-month periods are equivalent or not. The distribution of the radon concentration ratio during the second and the first period is shown in fig. 1 for both measured rooms. The ratio ranges from 0.6 to 1.8 for kitchens or living rooms, and from 0.6 to 1.3 for the bedrooms, with a median value of 0.9 for both room types. Therefore, the average radon concentration during the two exposure periods appears to be nearly equivalent, although with some house-to-house variation that will be discussed later.

The annual average values are also summarized in tab. 1. The Complete measurements were available for 17 dwellings. For these houses, the annual average for each room was obtained by weighting the results in the two periods, with the weights equal to the exposure times, in order to take into account minor deviations from the planned six months exposure period Then, the house annual average was obtained by averaging the annual averages of the two rooms. As regards the houses with missing measurements in one of the two six-month periods, it is possible to estimate the house annual average by using the ratio (evaluated in houses with complete measurements) of radon concentration in the two measuring periods. In particular, for each room type, the annual average was obtained multiplying the radon concentration in the first six-month period by the median values reported in fig. 1.

However, it is worth noting that the above procedure of estimating the annual average by using a correction factor based on the median of the ratio for the two periods introduces an uncertainty related to the width of the ratio distribution in the fig. 1. This is a common problem for every annual average obtained from the measurement periods shorter than one year [14].

The minimum annual indoor radon concentration is 41 Bq/m^3 and it was observed in Gračanica. The maximum value (378 Bq/m^3) was measured in Sušica and it is the only measured indoor radon concentration (roughly corresponding to 4%) that exceeds 300 Bq/m^3,

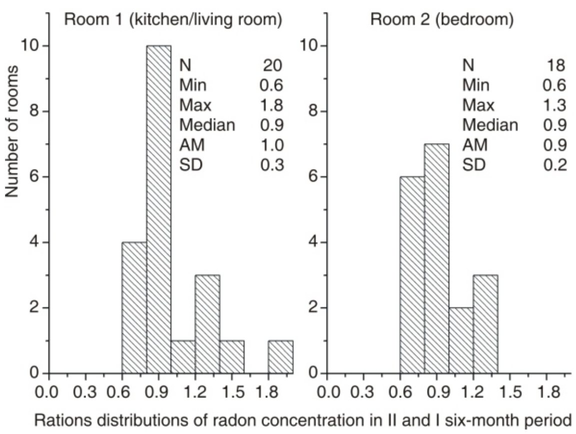

Figure 1. Ratio of radon concentration in the II six-month period and in the I six-month period for both measured room

which is the maximum reference level for existing houses, as established in the latest drafts of both the International and European Basic Safety Standards [15].

The results reported in tab. 1 can be compared with those of previous studies carried out partly in the same regions and partly in different regions of Kosovo. From measurements in 83 buildings in Priština [16], the results of indoor radon measurements ranging from 20 to 100 Bq/m^3 (with AM = 50 Bq/m^3) have been reported. The results of combined studies [17] are also quite different giving an approximate value of GM of 220 Bq/m^3. Part of the differences is probably due to the measuring period that was 3 months for previous studies and a total of 12 months in this study. The effect of the seasonal variations cannot be evaluated in this study because the chosen exposure periods were nearly equivalent as regards the average radon concentration. In [7] a radon values comparison in different periods was reported, showing higher values in winter than in summer, with ratios of average in different periods ranging from about 1 to about 1.8 (the comparison was done in different houses). In a study carried out in schools in Sharr municipality [5], very different average values were measured in winter (236 Bq/m^3) and summer (20 Bq/m^3), however these results could be not representative for dwellings, due to the peculiar characteristics and the occupancy of schools.

Activity concentration of natural radionuclides in soil

A total of 15 3 soil sample measurements were done.

The samples came from the undisturbed areas near the houses where also indoor radon concentration was measured. Table 2 gives the descriptive statistic for activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K for all samples obtained by gamma spectrometry measurements. The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K for all the 15 measuring sites collected at different depth are plotted in fig. 2. The plots show no significant increase of the

Table 2. Descriptive statistic of ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations in all the 15 3 soil samples

Radionuclide	Depth [cm]	Activity concentration [Bqkg^{-1}]				
		Min	Max	Med	AM	SD
^{226}Ra	0-5	16	45	23	24	8
	10-20	14	46	24	25	8
	20-40	16	59	22	27	11
^{232}Th	0-5	18	52	33	34	9
	10-20	18	52	33	36	9
	20-40	21	55	38	37	11
^{40}K	0-5	340	880	495	520	150
	10-20	292	912	506	529	157
	20-40	299	993	513	553	183

mean values increasing the depth, and therefore the values averaged over the three depths were used for the radiological impact estimations. The mean values of ^{226}Ra and ^{232}Th in the measured soil samples are slightly lower than the world population weighted mean values of 32 and 45 Bq/kg, respectively, whereas the measured mean value of ^{40}K is slightly higher than the world mean of 412 Bq/kg [18].

Correlation between the radium in soil and the indoor radon

In fig. 3 the radon indoor versus concentration of ^{226}Ra (averaged over the three depths) is plotted for each measurement site. The plot and the correlation parameter (R^2 = 0.260) show that the correlation is low. Therefore, we can conclude that the indoor radon levels in the selected dwellings are mostly affected by parameters other than the radium content in soil, such as other soil characteristics (*e. g.* permeability), house characteristics (particularly those related to the contact of the building with soil, but also wall materials), and living habits (*e. g.* ventilation).

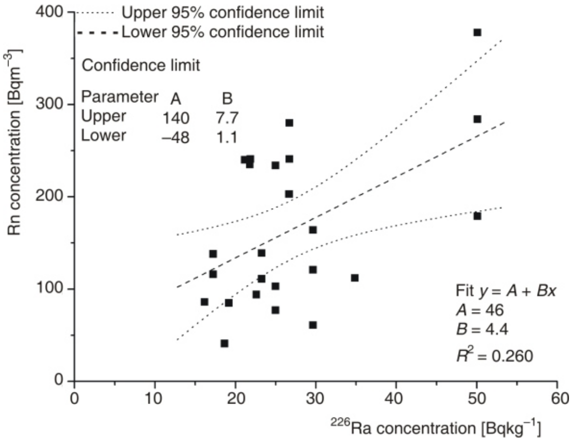

Figure 3. ^{226}Ra in soil *vs.* indoor radon concentration

Radiological impact

Radiological impact due to the indoor radon

The annual effective dose *E* [Sv] due to the exposure to indoor radon and its decay products can be

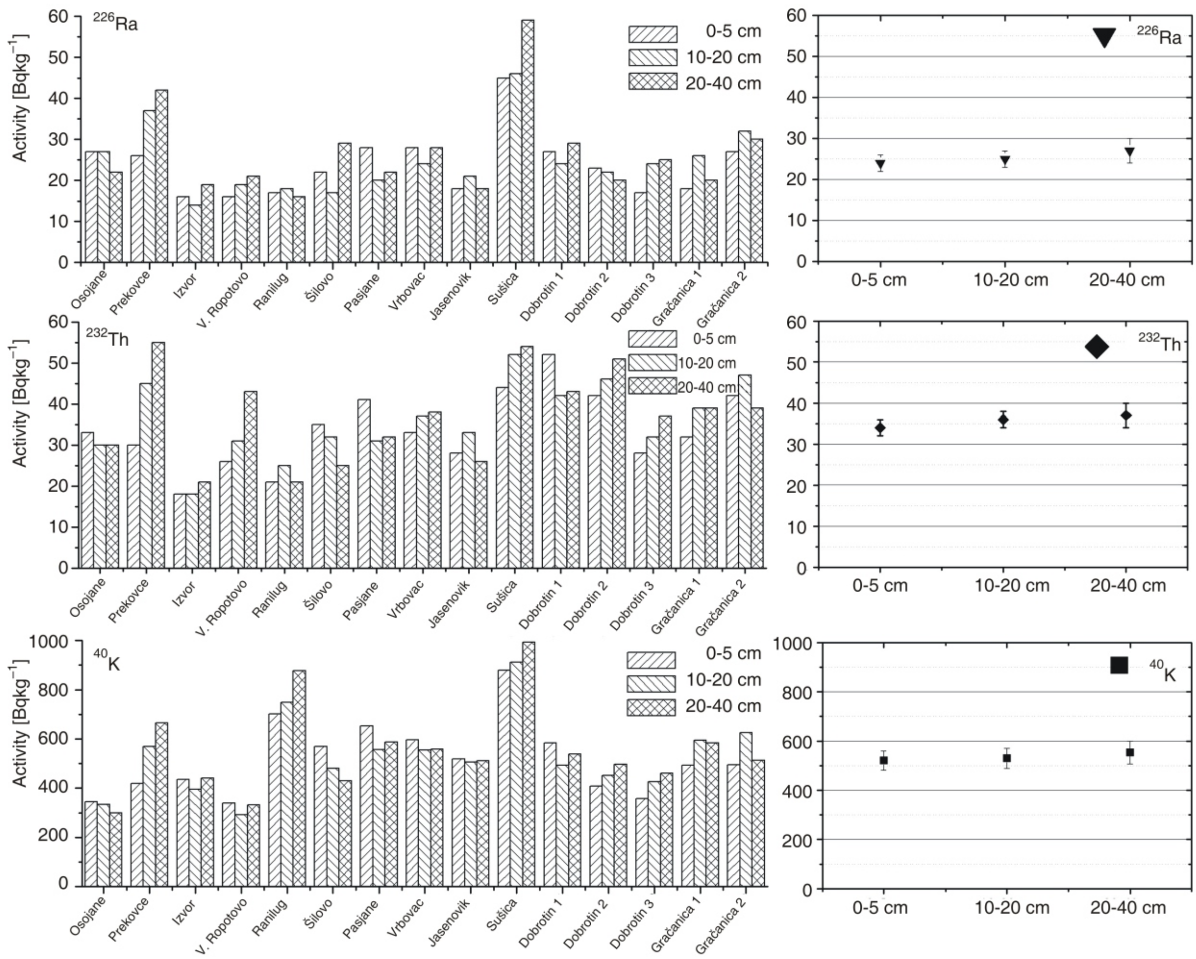

Figure 2. The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in soils sampled from different depth

estimated from the measured annual radon concentration using the following formula

$$E = C_{\mathrm{Rn}} F T DCF \qquad (1)$$

where C_{Rn} is the annual mean radon concentration, F – the equilibrium factor between radon and its decay products (taken equal to 0.4), T – the annual indoor occupancy (taken equal to 7000 hours per year as assumed also by ICRP [19]), and DCF – the dose conversion factor for radon decay products. Both the recently updated ICRP DCF and the usual UNSCEAR one – 12 and 9 nSv per Bqh/m^3, respectively – were considered [18, 19]. The annual effective dose values calculated by using the ICRP DCF is 5.5 mSv (calculated from the average indoor radon concentration in all 25 houses), are quite higher than the worldwide mean value of 1.2 mSv [19]. By using the UNSCEAR DCF, all annual effective doses resulted 3/4 lower than the above ones.

The mean value of effective dose obtained in this study by using ICRP DCF can be compared with results obtained in other studies in Balkan regions (tab. 3).

Radiological impact due to the outdoor terrestrial radiation

The outdoor exposure of general public and the corresponding absorbed dose rate and annual effective dose can be evaluated on the basis of the measured activity concentration in the soil. The absorbed dose rate D [nGyh^{-1}] in air at 1 m above the ground level due to the presence of natural radionuclides is estimated according to the following equation

$$D = 0.445\, A_{\mathrm{Ra}} + 0.584\, A_{\mathrm{Th}} + 0.0417\, A_{\mathrm{K}} \qquad (2)$$

where A_{Ra}, A_{Th}, and A_{K} are the average activity concentrations in Bqkg^{-1} of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The coefficients used in eq. (2) are obtained averaging the values found in literature, which range from 0.399 to 0.463 nGy/h per Bq/kg for ^{226}Ra, from 0.544 to 0.604 nGy/h per Bq/kg for ^{232}Th, and from 0.0399 to 0.0429 nGy/h per Bq/kg for ^{40}K [20-23]. The estimated average absorbed dose rates were found not to exceed the world average value of 60 nGy/h obtained from the direct outdoor measurements [19]. The absorbed dose rate values can be used to estimate an annual effective dose due to the gamma dose terrestrial radiation at each site. The conversion coefficient of 0.7 Sv/Gy from absorbed dose in air D [nGy] to effective dose E [μSv] and the outdoor occupancy factor of 0.2 for one year (8760 h) – 10^{-3} if E in μSv – proposed by [21], were used

$$E\,[\mathrm{mSv}] = D\,[\mathrm{nGy}]\; 1.23 \qquad (3)$$

In tab. 4 the summary of annual effective dose E statistic is presented. It can be seen that the outdoor annual effective dose ranges from 44 mSv (Izvor) to 111 mSv (Sušica), with an average of 66 mSv.

In tab. 5, a summary of average values of the annual effective doses due to outdoor exposure to natural radionuclides in soil is presented, as evaluated in this and other conducted investigations in Balkan region.

Table 4. Summary statistic of annual effective dose *E* in all the monitored locations

	AM	SD	Min	Max	Med	1stquart.	3rdquart.
E [Sv]	66	16	44	111	66	57	72

Table 5. Annual effective doses from natural radionuclides in soil, in Balkan region

Region or country	*E* [Sv]	Reference
Kosovo and Metohija	66	Present study
Republic of Srpska	85	[24]
Vojvodina	91	[25]
Serbia	77	[26]
FYR Macedonia	88	[27]

CONCLUSIONS

The annual average radon concentration measured in the selected dwellings distributed in 13 villages of 8 rural municipalities of South-Eastern and

Table 3. Indoor radon concentration and annual effective dose reported in different Balkan regions

Region or country	C_{Rn} [Bqm^{-3}]					E** (ICRP) [mSv]	Reference
	N	AM	(SD)	GM	(GSD*)	AM	
Kosovo and Metohija *(South-eastern and Central part)*	25	163	(84)	143	(1.7)	5.5	Present study
Serbia *(Niška Banja)*	65	1163		529	(3.9)	39	[17]
Kosovo and Metohija *(Central Kosovo, Region Prizren and North Kosovo)*	63	429		224	(3.3)	14	[6]
Montenegro *(Montenegrin coast)*	107	32	(23)	26	(2.1)	1.1	[2]
FYR Macedonia	437	105	(84)	84	(1.9)	3.5	[1]

* GSD is dimensionless
** All the annual effective dose values *E* are calculated by using the ICRP DCF

Central Kosovo and Metohija resulted quite high, although lower than values obtained during the three-month measurements in other areas of Kosovo and Metohija. On the opposite, the average of natural radionuclides activity concentration (and the derived doses) in soil resulted slightly lower (for ^{226}Ra and ^{232}Th) or slightly higher (for ^{40}K) than the world population weighted mean values. Although the highest indoor radon concentration and the highest radium activity concentration in soil were measured in the same location (Susica), the correlation between these two quantities is low and the indoor radon levels appearto be more affected by other parameters than the radium content in soil. Finally, the radon concentration was quite similar (on average) for the two selected six-month periods, so that a single six-month period (from December to June, or from June to December) could be used to obtain a good estimate of the annual average for a group of dwellings, whereas for a single dwelling this would introduce a significant uncertainty due to the house-to-house variability. This could be very useful in the design of large surveys.

ACKNOWLEDGEMENTS

The authors highly appreciate the kind co-operation of the owners of the houses where the detectors were deployed. The financial support is partially provided by P 41 028 of the Ministry of Education, Science and Technological Development of the Republic of Serbia.

AUTHOR CONTRIBUTIONS

Theoretical analysis was carried out by L. R. Gulan, F. Bochicchio, C. Carpentieri, G. A. Milić, Z. A. Stojanovska, D. R. Nikezić, and Z. S. Žunić. Experiments were organized and carried out by L. R. Gulan, C. Carpentieri, G. A. Milić, J. M. Stajić, D. Ž. Krstić, and Z. S. Žunić. All authors analyzed and discussed the results. The manuscript was mainly written by L. R. Gulan and F. Bochicchio, with the contribution of all the authors.

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Received on July 7, 2012
Accepted on January 28, 2013

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ВИСОКА ГОДИШЊА КОНЦЕНТРАЦИЈА РАДОНА У КУЋАМА СЕОСКИХ ПОДРУЧЈА КОСОВА И МЕТОХИЈЕ И ПРИРОДНА РАДИОАКТИВНОСТ ОКОЛНОГ ЗЕМЉИШТА

У претходним истраживањима концентрације радона у кућама (стамбеним просторијама) неких подручја Косова и Метохије измерене су високе просечне концентрације радона, премда су детектори били изложени само три месеца. Да би се могла планирати обимнија истраживања у овом подручју, годишња мерења у 25 кућа су спроведена као пилот студија. У свакој кући, у две просторије, дневној и спаваћој соби, постављени су пасивни радонски детектори Cr-39, сваки два пута по шест месеци у току годину дана, да би се видела и сезонска варијација. У циљу процене корелације измерених концентрација радона са садржајем радијума у околном земљишту и да би се унапредило знање о садржају природних радионуклида у земљишту, узорци земљишта су узорковани у околини сваке куће и анализирани на садржај радијума (^{226}Ra), торијума (^{232}Th), и калијума (^{40}K). Измерена просечна годишња концентрација радона од 163 Bq/m^3 је висока, а измерене концентрације у дневним и спаваћим собама се нису значајно разликовале међусобно као ни у првом и другом шестомесечном периоду у току годину дана, у току којих су детектори били излагани. У земљишту је расподела природних радионуклида била сасвим уједначена. Корелација између садржаја ^{226}Ra и концентрације радона у кућама била је ниска са коефицијентом корелације од (R^2 = 0.26). Годишња ефективна доза од радона и његових краткоживећих потомака (5.5 mSv, просечно) израчуната је коришћењем најновијих ICRP дозних конверзионих фактора. Допринос годишњој ефективној дози од спољашњег терестријалног гама зрачења које потиче од природних радионуклида је скоро занемарљив (66 μSv). Закључак је да су уочене високе концентрације радона само делимично корелисане са радијумом из земљишта. Добра процена годишње просечне концентрације радона може се добити на основу шестомесечног мерења са добро изабраним периодом излагања, што може бити корисно када се планирају обимнија истраживања.

Кључне речи: радон, радиоактивност тла, радиолошки утицај