PREDICTION OF LONG-TERM INDOOR RADON CONCENTRATION BASED ON SHORT-TERM MEASUREMENTS

by

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We present a method for the estimation of annual radon concentration based on short-term (three months) measurements. The study involves results from two independent sets of indoor radon concentration measurements performed in 16 cities of the Republic of Macedonia. The first data set contains winter and annual radon concentration obtained during the National survey in 2010 and the second, contains only the radon concentration measured during the winter of 2013. Both data sets pertain to radon concentration from the same cities and have been measured applying the same methodology in ground floor dwellings. The results appeared to be consistent and the dispersion of radon concentration was low. Linear regression analysis of the radon concentration measured in winter of 2010 and of the 2010 annual radon concentration revealed a high coefficient of determination $R^2 = 0.92$, with a relative uncertainty of 3%. Furthermore, this model was used to estimate the annual radon concentration solely from winter-term measurements performed in 2013.

The geometrical mean of the estimated annual radon concentration of the 2013: radon concentration (A-2013) =98 Bqm^{-3} was almost equal to the geometrical mean of the annual radon concentration from the 2010, radon concentration (A-2010) = 99 Bqm^{-3}. Analysis of the influence of building characteristics, such as presence/absence of a basement in the building, or the dominant building material on the estimated annual radon concentration is also reported. Our results show that a low number of relatively short-term radon measurements may produce a reasonable insight into a gross average obtained in a larger survey.

Key words: ground floor dwelling, indoor radon, linear regression analysis, uncertainty

INTRODUCTION

The majority of the urban population spends long periods indoors, where radon accumulates, which may lead to elevated indoor concentrations. Radon is known as the most significant contributor to the dose received by the population due to exposure to natural sources of ionizing radiation [1]. To reduce the risk from radon exposure, authorities of many countries prepared national radon programmes the overall designs of which are based on the experience gained from the national surveys. The programmes incorporated the knowledge about the temporal and spatial variability of the radon concentrations (C_{Rn}) controlled by numerous natural and anthropogenic factors [2, 3]. A radon survey qualifies as being national when it involves simultaneously measuring the C_{Rn} in a representative sample of dwellings throughout the entire country during a year. In general, the measurements are performed with nuclear track detectors.

The detectors are usually deployed for the following periods of time:

(a) quarterly, where the detectors are exposed in four successive periods of three months (one full season) [4-6],
(b) semi-annually, two successive periods for a duration of six months, including two full seasons [7], or, and

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(d) annually, for a period of one year [8, 9].

Regardless of the time of exposure, the results are expressed as an annual C_{Rn}. In cases (a) and (b) the annual C_{Rn} is presented as arithmetic mean of the measured concentrations in the successive periods. In the case under (c) the annual C_{Rn} is measured directly.

The radon problem became a serious concern throughout the Balkan region. Large amounts of regional data became available from recent surveys carried out in Serbia [10-14], the Republic of Srpska [15, 16], Bulgaria [17], Romania [18] and Greece [19]. Also, several campaigns of C_{Rn} measuring have been conducted in the Republic of Macedonia over the last decade. The greatest source of data was provided by the national surveys of radon and thoron in dwellings in the Republic of Macedonia from 2010 [20-22]. Further research of C_{Rn} was conducted in schools and dwellings [6, 8]. Moreover, some investigations were done for establishing a relationship between the indoor C_{Rn} and the activity of ^{226}Ra in surface soil [23]. As part of these activities, a supplementary survey was carried out in 47 dwellings from 16 cities during the winter months of 2013. The results are a subject of this study. We present the evaluation of annual C_{Rn} and its uncertainty comparing the variance of results with those of the national survey measured in the same cities in 2010. Variation related to building characteristic is also discussed.

MATERIALS AND METHODS

Design of survey

The radon detectors were distributed within a campaing that involved undergraduate students from the first year of the Faculty of Medical Sciences in Stip, class 2012/2013. Students originating from various cities throughout the country were instructed to deploy a radon track-detector at the ground floor of their homes in their home cities. They were asked to keep the detector deployed during a three-month winter period (January to March 2013). The 47 detectors were deployed at a distance greater than 50 cm away from a wall and away from a heating source in the most occupied room (either living room or bedroom). The students filled out a questionnaire, providing general information about the characteristics of the house or building: position (GPS co-ordinates), presence/absence of basement, smoking habits, and the type of dominant building material. Some of these factors were further considered as a grouping parameter in the statistical analysis of the results. Four of the detectors were lost during the campaign. Finally, we received results for 43 dwellings located in 16 cities, pertaining to winter 2013. The cities under observation are characterized by different geographical position trough the country (fig. 1), and are located at altitudes between

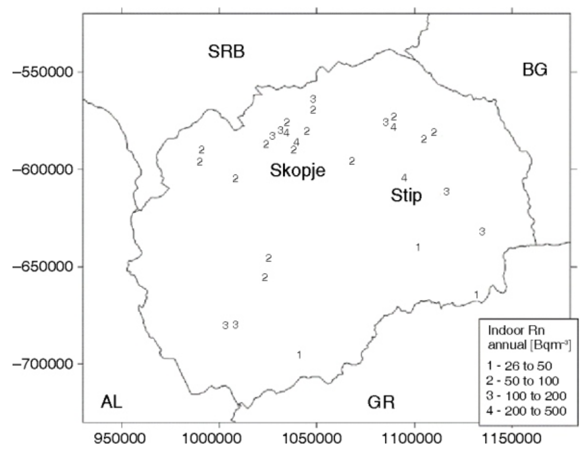

Figure 1. Spatial estimate of annual C_{Rn} in 43 dwellings; coordinates in m, GISCO Lambert azimuthal equal area projection

70 m to 650 m above sea level. They also belong to different geological zones categorised in different litho-stratigraphic units.

Using the list of the cities included in the 2013 survey we extracted the C_{Rn} results for the winter season in 2010, C_{Rn}(W-2010), and for the entire year 2010, C_{Rn}(A-2010). The 2010 dataset included C_{Rn} results from 186 different dwellings in 15 cities. One of the cities from the 2013 survey was not included in the list of the survey in 2010, but was kept for the study.

Measurements

The radon concentration in 2013 was measured using the same nuclear track detectors with commercial name RSKS, product of Radosys, Hungary, that were used for the survey in 2010. The track detectors consisted of a CR-39 detector placed in a cylindrical diffusion chamber with 25 mm 40 mm. After being collected, the detectors were sent to the National Centre of Radiobiology and Radiation Protection in Sofia Bulgaria, for analysis.

The CR-39 detectors were detached from the diffusion chambers, and were chemically etched in 6.25 M solution of NaOH at a temperature of 95 °C for 3.4 hours. The track counting was performed by the optical transmission microscope using an automated image analysis system. The software identified the detector's ID code engraved onto the surface of the CR-39. Using an appropriate calibration factor, the software converted the detected number of tracks per unit area (track density) into radon concentration, expressed as

$$C_{Rn} = f_c \frac{\rho - \rho_b}{t} = f_c \frac{\rho_{net}}{t} \tag{1}$$

were C_{Rn} is radon concentration (in kBqm^{-3}), while ρ_b and ρ are the background and counted track density per mm^2, respectively; f_c is a calibration factor, and t is the exposure time in hours. The calibration factor for this series of detectors was provided by the manufacturer, $f_c = 44.47 \pm 3.53$ (in tracks per mm^2/ kBqh per mm^{-2}). The background track density $\rho_b = 0.417 \pm 0.05$ (in tracks per mm^2) was determined from 10 blank detectors measurements.

To determine the annual radon concentrations by using short-term (winter) measurements one should first establish the relation between them. For this purpose we have provided a linear regression analysis of the measured C_{Rn} in the winter 2010, C_{Rn}(W-2010) and the annual C_{Rn}(A-2010). The analysis yielded the following eq.

$$C_{Rn}(A) = aC_{Rn}(W) + b = af_c \frac{\rho_{nst}}{t} + b \qquad (2)$$

where *a* and *b* are the constants of the linear dependence of the annual *vs*. the winter C_{Rn}.

The quantification of the uncertainty was done in accordance with the method, recommended in the EURACHEM Guide [24] and IAEA Tec doc 1401 [25]. The combined standard uncertainty was estimated by applying the law of uncertainty propagation for the independent variables

$$u_c(y) = y\sqrt{\left(\frac{u(x_1)}{x_1}\right)^2 + \left(\frac{u(x_2)}{x_2}\right)^2 + \ldots + \left(\frac{u(x_n)}{x_n}\right)^2} \qquad (3)$$

where the $u(x)/x$ are the uncertainties expressed as relative standard deviations.

The uncertainty of the annual radon concentration was estimated taking into account its probability distribution, converting each source of uncertainty into a standard uncertainty, and finally, combine them, as shown in eq. 4.

According to eqs. 2 and 3, the combined uncertainty of the estimated annual concentration C_{Rn} (A) is given in eq. 4

$$u_c[C_{Rn}(A)] = C_{Rn}(A)\sqrt{\left(\frac{u(f_c)}{f_c}\right)^2 + \left(\frac{u(\rho_{net})}{\rho_{net}}\right)^2 + \left(\frac{u(t)}{t}\right)^2 + \left(\frac{u(a)}{a}\right)^2} \qquad (4)$$

The uncertainty of the calibration factor $u(f_c)$ was provided by the manufacturer (Radosys). Its relative value was 7.9 %.

The uncertainty of the net track density is calculated as a combined uncertainty of the track density and the background track density of an unexposed (blank detector), as given with eq. 5

$$u(\rho_{net}) = \rho_{net}\sqrt{\left(\frac{u(\rho)}{\rho}\right)^2 + \left(\frac{u(\rho_B)}{\rho_B}\right)^2} \qquad (5)$$

Both uncertainty components in eq. 5 were determined from multiple series of measurements. Repeated measurements of detectors for QA with low, medium, and high track density gave the relative uncertainties of 8, 5, and 3 %, respectively. The relative uncertainty of 12.5 % for the background uncertainty was obtained from repeated measurements of 10 blank detectors.

It should be also noted that the exposure time in eqs. 1 and 2 is expressed in hours. On the other hand, the reported time of detector exposure is measured in days (24 h). Therefore, we assume a possible contribution of several hours' difference at the times of deployment of 46 track-detectors to the overall uncertainty. Lacking the knowledge about the shape of the data distribution, but knowing the time range of ±2 day (1 day for deploying and 1 day for collecting detectors), we have estimated this type of uncertainty theoretically. Assuming a rectangular distribution of the exposure time, the uncertainty was calculated as $u(t) = 2/\sqrt{3}$ in days. For the detectors exposed during three months in winter 2013, the relative uncertainty for a time exposure was within the range: from 0.8 to 1.7 %.

The uncertainty of the constant *a* in eq. 4 was expressed with the uncertainty originating from the linear least squares fitting procedure. It was calculated as the root mean square (RMS) of the differences between the experimental and modelled values. It was found to be 3 %.

Finally, in this study, the relative combined uncertainty of annual C_{Rn}, that included the uncertainties of calibration factor, detector background, track counting, exposure time and seasonal correction was found to be between 10 % and 14 %.

RESULTS AND DISCUSSION

Table 1 shows the descriptive statistics of the indoor C_{Rn}. The last two columns refer to the results of the survey from 2010, whereas the second last column refers to winter quarterly measured C_{Rn}(W-2010) and the last one corresponding to the annual radon concentration C_{Rn}(A-2010), that was calculated as an average C_{Rn} from the measurements of the four seasons in 2010. The first and the second column pertain to the data from 2013, where C_{Rn}(W-2013) represents the measured values in the winter and C_{Rn}(A-2013) is the estimated annual value with applied seasonal correction, using the linear model from 2010. The measuring locations and the estimated annual C_{Rn}(A-2013) are mapped on fig. 1.

The results were fitted with a log-normal function. Kolmogorov-Smirnov and Chi-square tests were used for testing the hypothesis that the data follow a log-normal distribution. For all data sets, the assumption was confirmed at 95 % level of significance. Furthermore, normality was tested by using the Anderson-Darling and Shapiro-Wilk tests. The summary results of the tests are presented in tab. 2. The obtained values for the error probability (*p*) were greater than 0.05, and thus confirmed the hypothesis for log-normality and normality.

Table 1. Descriptive statistics of indoor radon concentrations covered in this work

Statistic	C_{Rn}(W-2013) [Bqm^{-3}]	C_{Rn}(A-2013) [Bqm^{-3}]	C_{Rn}(W-2010) [Bqm^{-3}]	C_{Rn}(A-2010) [Bqm^{-3}]
Number of observations	43	43	186	186
Minimum	30	26	17	18
Maximum	535	460	956	552
Median	110	95	135	96
Mean	140	120	179	123
Standard deviation	98	85	150	93
Variation coefficient (CV)	70 %	69 %	84 %	75 %
Geometric mean (GM)	114	98	137	99
Geometric standard deviation (GSD)	1.90	1.90	2.06	1.91

Table 2. The summary results of distribution fitting tests

Variable/test	Kolmogorov-Smirnov	Chi-square	Shapiro-Wilk	Anderson-Darling
	Log-normality testing original data		Normality testing ln transformed	
C_{Rn}(W-2013)	$p = 0.998$	$p = 0.454$	$p = 0.996$	$p = 0.992$
C_{Rn}(A-2013)	$p = 0.998$	$p = 0.421$	$p = 0.996$	$p = 0.992$
C_{Rn}(W-2010)	$p = 0.853$	$p = 0.033$	$p = 0.762$	$p = 0.553$
C_{Rn}(A-2010)	$p = 0.719$	$p = 0.150$	$p = 0.364$	$p = 0.317$

Linear model development

The model was developed with the parametric linear regression analysis applied to the results from 2010. It appeared that the dependence among the annual concentrations of radon can be very well described with a linear function. The results are shown graphically in fig. 2. The regression coefficients of the linear model, $C_{Rn}(A) = C_{Rn}(W)\,a + b$, are as follows: $a = 0.859$ and $b = 0.369$, with a high coefficient of determination $R^2 = 0.92$.

Comparison between C_{Rn} measured in 2010 and 2013

We compared the homogeneity of the C_{Rn}(2010) and C_{Rn}(2013) variances using the Bartlett test (BT). The null hypothesis assuming the variables have the same variance, was confirmed for p 0.05. From here it follows that the variances of the measured C_{Rn}(W-2010) and C_{Rn}(W-2013) were practically equal (BT, $p = 0.978$). Likewise, it appeared that the variance of the annual concentrations C_{Rn}(A-2010) and C_{Rn}(A-2013) were also equal (BT, $p = 0.952$).

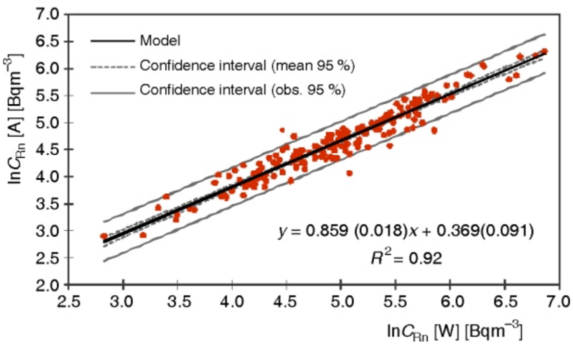

Figure 2. Result of the regression analysis of annual and C_{Rn} measured in the winter of 2010. Regression model: $y = ax + b$ (in brackets: uncertainty of coefficient)

The differences between GM values were also tested. Since the data followed the normal distribution and the variances of the two data sets were equal, the Student's *t*-test for independent samples/two-tailed, was applied for the log-transformed values of the C_{Rn}. The differences between the GM values of the measured winter C_{Rn} for both surveys (2010 and 2013) were negligible (Student *t*-test, $p = 0.124$). It also applies to the differences between the estimated annual mean values (Student *t*-test, $p = 0.924$).

Impact of the building characteristics

We also compared the variances of C_{Rn} for two groups of data, classified according to either the presence or absence of a basement in a dwelling. Furthermore, we compared the variances between the groups of houses that were made of different dominant construction material. The results of the Bartlett test for all the groups are given in tab. 3. Because the datasets are homogeneous (BT, $p > 0.05$) and normal (AD, SW, $p > 0.05$), the influence of these factors was tested on the grouped C_{Rn} by ANOVA (analysis of variance) and Fisher LSD test.

ANOVA for winter and annual CRn from 2013, grouped according to the "presence of basement" and "building materials" showed significant differences between the mean values of the groups (ANOVA, $p < 0.0001$ in both cases). The 2010 data test also revealed significant differences. ANOVA gave a value $p = 0.006$ for the winter measurements and $p = 0.001$ for the annual values, respectively.

The Fisher LSD test for C_{Rn} winter measurements showed that its mean radon concentration values in houses without basements are higher than those

Table 3. Results of the grouped C_{Rn} by presence of basement and building materials homogeneity testing

	Bartelett test	
	Grouped by basement	Grouped by bulding materials
ln C_{Rn} (W-2013)	0.493	0.922
ln C_{Rn} (A-2013)	0.485	0.924
ln C_{Rn} (W-2010)	0.874	0.403
ln C_{Rn} (A-2010)	0.449	0.294

measured in houses with basements, p = 0.0002 (fig. 3). Apparently, the results showed simmilar behaviour for the annual concentrations of both surveys. The mean value for C_{Rn} measured during the winter season of 2010 is LSD, p = 0.001 and for the annual value LSD, p = 0.0003, respectively.

The influence of building materials on the indoor C_{Rn} is not as obvious as the one of the presence of a basement, as was previously described by other authors [26, 27]. For example, in the 2013 survey, the impact of building materials is significant (LSD, p = = 0.006 (winter); p = 0.006 (annual)) while in the 2010 survey it is not (LSD test, p = 0.261 (winter); p = 0.174 (annual)). Figure 4 shows the GM values of C_{Rn} taken from both surveys (2010 and 2013), grouped with respect to the type of building material. The mean value of C_{Rn} measured in winter in the new survey for houses built of concrete was lower than for those made of stone (LSD, p = 0.003) and lower than for those constructed of brick (LSD, p = 0.009). The mean values were divided into two groups: Group 1 – houses built of brick and stone and Group 2 – houses built of concrete. Figure 4 reveals that a similar trend exists for the values from the national survey but it is not represented by a significant grouping as in the new survey. For example, the differences were significant only between the mean C_{Rn} for the houses made of stone and concrete (LSD, p = 0.037 for the measurements in winter), (LSD, p = 0.021 for annual concentration). However, the differences between concentrations related to houses of bricks and houses of stone were not significant. Hence, the C_{Rn} values can be grouped in two groups (brick/stone and concrete).

To summarize, it should be noted that: the factor "basement" clearly showed the influence of geogenic radon on indoor C_{Rn}. On the other hand, building material appeared to be a less dominant factor that influences the indoor C_{Rn}.

Figure 5 was created for further clarification of the influencing factors. It shows the geometric means (GM) of C_{Rn} for both surveys grouped by two factors: the presence of basement and building materials. For example, in houses with no basement (1 and 2 bars) for which the dominant building material is brick, there is no diference between the 2010 and 2013 survey (error bars overlapping). However, notable differences appeared for brick houses with basements (error of 3 and 4 bars overlapping). That this may be affected by the season factor is indicated by the fact that this difference does not appear for annual C_{Rn} in houses made of bricks with basements. In the same figure, the seasonal impact can be seen in houses with basements built of concrete. For houses made of stone with no basement, the results cannot be compared because the 2013 survey includes only one such house. However, one can see that the error bars of radon concentration in all groups overlap, meaning that all groups have the same mean values.

Furthermore, the results of C_{Rn} measured in the buildings made of concrete showed slightly different behavior than brick and stone houses. The C_{Rn} values for 2013 pertaining to the group of houses built of concrete are lower than those from the 2010 survey measurements (the 1 and 3 bars are lower than the 2 and 4 ones in fig. 5). The apparent inconsistencies can be explained by a small number of samples in 2013, as well as that the samples do not necessarily represent the population exposed to the same environment, as the geology of cities may vary. Furthermore, the house characteristics are determined by many other factors other than the presence of basement and building material, which cannot be expected to be "averaged

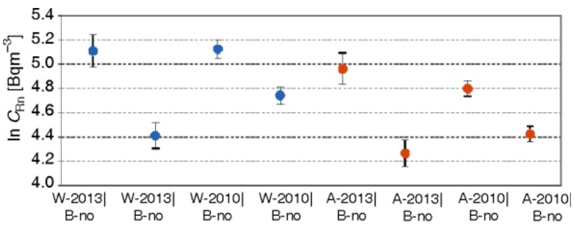

Figure 3. Geometric means of C_{Rn} in houses with and without basements

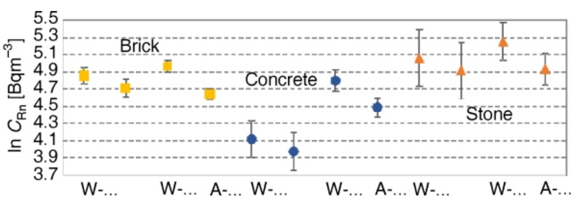

Figure 4. Geometric means of C_{Rn} in houses built from bricks, concrete and stone

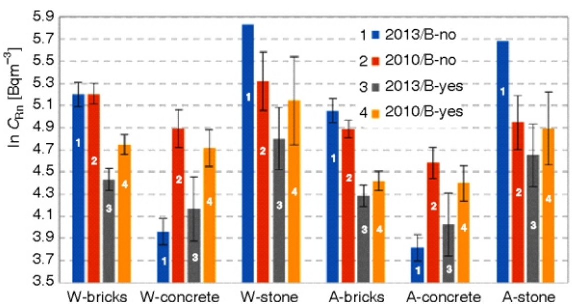

Figure 5. Geometric means of C_{Rn} in houses built of brick, concrete and stone in houses with and without basements

away" with only few samples. This illustrates that apart from the seasonal variations, the influence of some other factors related to characteristics such as age of the building [28, 29], type of windows [30], heating method [31], wall finishing [32], number of floors [33] and geology [34-36] should not be ignored.

CONCLUDING REMARKS

A student campaign was organized in the winter of 2013 for indoor C_{Rn} measurements in ground floor rooms of 43 dwellings in 16 randomly selected cities in the Republic of Macedonia.

To estimate the annual C_{Rn} for 2013, the winter data were adjusted to annual means by applying a linear model, derived from the results of the 2010 survey by regression of annual *vs*. winter. The obtained function in this way has a high coefficient of determination and a negligible contribution to the combined uncertainty of the annual radon concentration.

Analysis of the data shows that C_{Rn} measured in the winter as well as estimated annual C_{Rn} from the 2013 and 2010 survey:
- can be described with log-normal distributions,
- have the same variance and GM values (insignificant difference),
- have the same variances when grouped with respect to the presence of basement and the dominant building material,
- showed the same trend of the differences in GM values between C_{Rn} in houses with and with no basement, and
- showed the same trend of differences of GM values between the C_{Rn} values for houses built from concrete and stone.

The proposed method aims to provide time and cost effective measurements (three-month duration – single disposable detector) for estimation of the annual indoor radon concentrations C_{Rn}(A). Similar models could be developed for estimation of the annual C_{Rn} using a different season of the year (summer, spring or fall). Actually, something very similar has been done already in Macedonia [22] and India [37] where the method was used to estimate missing seasons.

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AUTHORS' CONTRIBUTIONS

The manuscript was written by Z. Stojanovska with contribution of all the authors. The figures were prepared by Z. Stojanovska and P. Bossew. The survey was organized by Z. Stojanovska and M. Zdravkovska and detector analysis were carried out by K. Ivanova and M. Tsenova. Theoretical investigation was carried out by Z. Stojanovska, K. Ivanova, P. Bossew, B. Boev, Z. S. Žunić, Z. Ćurguz, P. Kolarž, M. Ristova. Results discussion and review of the manuscript involved all authors.

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ПРОЦЕНА ДУГОТРАЈНИХ КОНЦЕНТРАЦИЈА РАДОНА У ЗАТВОРЕНИМ ПРОСТОРИЈАМА НА ОСНОВУ КРАТКОРТАЈНИХ МЕРЕЊА

Представљена је метода процене годишње концентрације радона заснован на краткотрајним (тромесечним) мерењима. Студија обухвата резултате два независна скупа мерења концентрације радона у затвореним просторијама спроведених у 16 градова Републике Македоније. Први сет резултата садржи зимске и годишње концентрације радона добијене током националног мерења радона у 2010. години. Други сет података садржи само концентраије радона мерене током зиме 2013. године. Оба сета података односе се на концентрације радона из истих градова, где су мерења вршена у становима и приземљу истом методологијом. Добијени резултати били су увек са ниском дисперзијом концентрације радона. Линеарна регресиона анализа примењена на концентрације радона измерене у току зиме 2010. и годишње концентрације радона за 2010. годину показала је висок коефицијент детерминације $R^2 = 0.92$, са релативном несигурношћу од 3 %. Овај линерани модел коришћен је за процену годишње концентрације радона за 2013. годину на основу зимских мерења изведених те године.

Геометријска средња вредност процењене годишње концентрације радона за 2013, (A-2013) = 98 $\mathrm{Bqm^{-3}}$, била је готово једнака вредности која се односи на концентрацију радона у 2010. години, (A-2010) = 99 $\mathrm{Bqm^{-3}}$. Анализа утицаја карактеристике зграда, одсуство подрума, односно доминантног грађевинског материјала, на процењене годишње концентрације радона, такође је приказана. Наши резултати показују да мали број релативно кратких мерења радона могу дати разуман увид у просечне резултате добијене у опсежнијем истраживању.

Кључне речи: стан у приземљу, радон у затвореној просторији, линеарна регресиона анализа, несигурност