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## ANNUAL AND SEASONAL VARIATIONS OF INDOOR RADON CONCENTRATION IN SKOPJE (REPUBLIC OF MACEDONIA)

Stojanovska Zdenka<sup>1,\*</sup>, Boev Blazo<sup>2</sup>, Ristova Mimoza<sup>3</sup>, Žunić S. Zora<sup>4,</sup> Zoran Curguz<sup>5</sup> Januseski Jovan<sup>6</sup>

<sup>1,4</sup> Faculty of Medical Sciences, Goce Delcev University, Stip, Republic of Macedonia
 <sup>2</sup>Faculty of Mining, Geology and Polytechnic, Goce Delcev University, Stip, Republic of Macedonia
 <sup>3</sup> Institute of Physic, Faculty of Natural Sciences and Mathematics, University in Skopje, Republic of Macedonia
 <sup>4</sup>ECE Lab, Institute of nuclear sciences "Vinča", University of Belgrade, Serbia
 <sup>5</sup>Faculty of transport Doboj, University in East Sarajevo, Republic of Srpska
 <sup>6</sup>Institute for Public Health of RM, Laboratory for radioecology Skopje, Republic of Macedonia

Abstract. This paper presents the results of a survey of indoor radon concentrations in the dwellings of 10 Municipalities in the Skopje: the capital and the largest city of the Republic of Macedonia. The indoor radon concentrations were measured during the four successive seasons from December 2008 to December 2009 using integrating passive alpha track-etch detectors with an exposure period of three months. The annual mean indoor radon concentration in each measuring site was estimated as an arithmetic mean from the four individual measurements

The measurements were completed for 124 dwellings, of which 112 dwellings revealed indoor radon concentrations lower than 200 Bq m-3, and 3 showed radon concentrations in excess of 400 Bq m-3. The annual mean indoor radon concentrations were found to be log-normally distributed, ranging from 18 to 502 Bq m-3. The geometric mean value of the indoor radon concentration in Skopje region was estimated to be 83\*/1.94 Bq m-3. The results of analysis of variance showed statistically significant differences in annual mean indoor radon concentrations among the different municipalities (p=0.021).

The influence of the factors linked to building characteristics in relation to the annual mean of indoor radon concentration was also a subject to examination. The factors which allow differentiation into subgroups (significance level p<0.05) were: the floor level (p<0.0001), presence of basement (p<0.0001), and type of heating (p=0.004). Seasonal dependence of indoor radon concentration was observed. The minimum indoor radon concentrations were found in the summer season whereas maximum levels were observed in the winter season (p<0.0001).

Key words: indoor radon, variation, seasonal variation

#### 1. Introduction

Concerning the radiation protection, the most dominant natural radiation source for the public exposure is the radon 222Rn and its short lived progeny, which occupies more than half of the dose exposure reaching the general public (1). Since indoor radon according to the World Health Organization is the second risk factor after smoking (2, 3), considerable effort has been performing in measuring, modeling and predicting such levels, remedying them were possible and reasonable (4). The pathway for radon generation in rock and soil to its indoors accumulation is controlled by a number of geogenic and anthropogenic factors, which leads to a large temporal and spatial variability (5, 6, 7). The seasonal variability of the indoor radon on the territory of Republic of Macedonia, investigated by Stojanovska et all, confirmed its regional distribution character (8). Furthermore, geological diversity of the Country contributes to the large scale of spatial variations (9).

In order to clarify variation and factors which enable to affecting the indoor radon concentration the results of indoor radon concentration, measured in Municipalities of Skopje, are analyzed observed and discussed.

### 1.1. Study area

Skopje is the capital and largest city of the Republic of Macedonia located in the northern part of the Country, in the center of the Balkan Peninsula, covering an area of 571.46 km². According to State Statistical Office, the territory is organized into ten municipalities, with a total population of 506 926(10). The center of Skopje is located at an elevation of 225 m with highest altitude of 1066 m above sea level.

With respect to the geology, Skopje belongs to the geotectonic Vardar zone, which is mainly composed of Neogene-Quaternary sediments, represented mainly by clay and sandstone (11). Skopje lies over a weak, highly porous zone, also associated with high seismic activity in that region (12). Its surface is formed at the intersection

between NW-SE and E-W trending deep Mesozoic magmatic structures (12).

The summers are hot and humid, while the winters are cold, wet, and often snowy. In summer, maximum daily temperatures are usually above 31 °C and sometimes above 40 °C. In spring and autumn, the temperatures range from 15 to 24 °C. In winter, the day temperatures are roughly 6 °C, whereas during the nights they often fall below 0 °C and sometimes below –10 °C. Occurrences of precipitation are evenly distributed throughout the year, being heaviest from October to December and from April to June.



Fig. 1 Skopje municipalities: Aerodrom (1), Butel (2), Cair (3), Centar (4), Gazi Baba (5), Gjorce Petrov (6), Karpos (7), Kisela Voda (8), Saraj (9), Suto Orizari (10)

#### 2. MATERIALS AND METHODS

Indoor radon concentrations were measured in 124 randomly selected dwellings through the entire Skopje area. In each dwelling, a detector was placed either in a bedroom or a living room, depending on where residents spent most of their time, at a height of 1 to 1.5 m above the floor, at a distance greater than 0.5 m from each wall, and at a minimum of 20 cm from any other object.

The commercially available RSKS and RADUET passive track detectors, produced by Radosys (Hungary), were used for measurements. RSKS detector consisted of CR-39 chip placed in a Ø25mm x 40mm cylindrical diffusion chamber. The RADUET detector was used for simultaneous detection of radon and thoron activity. It consisted of two detector chips CR 39, fixed in the pot section of two diffusion chambers (Ø60mm x 30mm). The main chamber was sensitive to radon, whereas the secondary chamber was sensitive to both, radon and thoron.

The detectors were exposed in the following periods:
December 2008 - February 2009 (winter period), March

– May, 2009 (spring period); June – August, 2009
(summer period) and September – November, 2009
(autumn period), using a different detector for each
period. After exposures of 3 months, the detectors were
detached, etched and scanned in the laboratory. The

recorded track density was then converted to Becquerel per cubic meter by using appropriate calibration factors.

The evaluation of the results was done using commercially available XLSTAT Pro 7.5 Statistic Software.

#### 3. RESULTS AND DISCUSSION

The histogram of annual mean indoor radon concentration obtained for 124 dwellings is presented on Figure 2. The obtained results are ranged from 18 to 502 Bq m<sup>-3</sup> of which 112 dwellings revealed indoor radon concentrations lower than 200 Bq m<sup>-3</sup>.

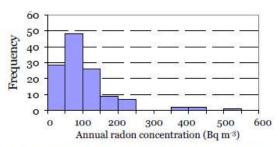


Fig. 2 Histogram of the annual indoor radon concentration

The annual indoor radon data set as a whole, like the sub-set grouped by season or municipalities appears lognormally distributed. The log-normality was tested by  $\chi_2$  and Kolmogorov-Smirnov tests, which showed, basically, the same results at a significance level of 95 % (p  $\leq$  0.05). The estimated parameters of the log-normal distribution are  $\mu$ =4.42 and  $\sigma$ =0.66, that is GM=83 Bq m<sup>-3</sup> and GSD=1.94. The arithmetic (AM) and geometric mean (GM) values with its standard deviation (GM) and geometric standard deviation (GSD) of annual radon concentrations well as the number of measured dwellings for each Municipality are reported in Table 2.

Table 2. Basic statistics of measuring results

Municipality	N	AM Bqm-3	SD Bqm <sup>-3</sup>	GM Bqm <sup>-3</sup>	GSD
Aerodrom	13	122	125	85	2.30
Butel	10	124	50	114	1.57
Cair	2	80	22	79	1.33
Centar	21	75	42	66	1.71
Gazi Baba	13	181	128	146	1.97
Gjorce Petrov	12	101	66	83	1.92
Karpos	28	85	59	69	1.93
Kisela voda	18	95	92	74	1.93
Saraj	2	85	33	82	1.49
Suto Orizari	5	125	41	120	1.33
Skopje	124	105	83	83	1.94

Since log transformed data have a normal distribution and the variance is homogeneous, the parametric analysis of variance (ANOVA) and Fisher's LSD-test was applied at a significance level of 95 % (p  $\leq$  0.05). LSD values were used for grouping regions

according to their mean values. In the following all differences are significant with p<0.05.

The municipalities geometric mean values are significantly different (ANOVA, p=0.021). The annual indoor radon concentration was found to be higher in Gazi Baba than for Centar (p=0.001), Karpos (p=0.001), Kisela Voda (p=0.003), Gorce Petrov (p=0.027) and Aerodrom (p=0.031) as well as the higher values in Butel than Centar (p=0.027) and Karpos (p=0.033) were found to be

However, in four municipalities (Aerodrom, Butel, Gazi Baba, Suto Orizari) the geometric mean values of the indoor radon concentrations were higher than national average of 84 Bq m-3 obtained from the National Survey (8).

In order to investigate the influence of the building characteristics to the annual indoor radon concentrations, the following factors were taken into consideration: house age, type of room, type of heating, floor level, presence of basement, buildings materials and type of windows. The factors which enable differentiation into subgroups (ANOVA, p<0.05) were found to be: the floor level (p<0.0001), presence of basement (p<0.0001), and type of heating (p=0.004). The graph on the Figure 3 confirms the fact that the indoor radon concentration decreases with floor level (the second floor is excluded from the analysis because of the low number of cases). On the same graph it is obvious that the influence presence/absence of basement is significant only for the ground floor values. Possible explanation of this finding could be the fact that the indoor radon concentration is mainly depending of the underlying soil characteristics.

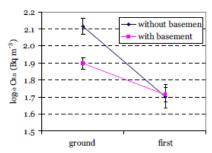


Fig. 3 Annual indoor radon concentration in the houses with and without basement as a function of floor level

Furthermore, influence of the heating system in the house on the indoor radon was also examined. In this subgroup we found statistically significant evidence of higher indoor radon concentration in houses with solid fuel heating in comparison with houses with central/electrical heating (Figure 4). For the first floor in houses with basement no significant influence was found because of the well-known stack-effect.

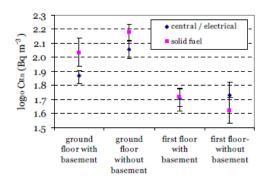


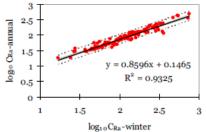
Fig. 4 Influence of type of heating on indoor radon concentration, measured as function of floor level and presence or absence of basement

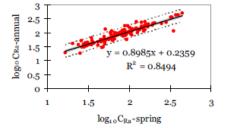
Analysis performed on the other factors as type of windows, building materials, house age and type of floor material. These resulted in no association with indoor radon concentration, but chance of its influence cannot exclude. However, the limited number of dwellings in this survey did not allow us to determine subgroups of adequate size for suitable investigation.

#### 3.1. Seasonal variation of indoor radon

It appeared that within each season, the measured indoor radon concentration follows a log-normal distribution.

In order to derivate seasonal correction factors for Skopje, the pattern of seasonal variation of indoor radon concentration was analysed using the data of the seasonal measurements in all dwellings. It was found that a liner function best fits the correlation of annual indoor radon concentration and indoor radon seasonal measurements. The results of regression analysis are presented in Figure 5.





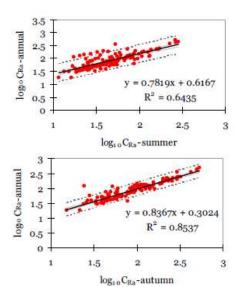


Fig. 5 Linear regression of log<sub>10</sub>C<sub>Ra</sub> by: log<sub>10</sub>C<sub>Ra</sub>(winter), log<sub>10</sub>C<sub>Ra</sub>(spring), log<sub>10</sub>C<sub>Ra</sub>(summer), log<sub>10</sub>C<sub>Ra</sub>(autumn),

Practically the same situation as on the national level, the highest correlation coefficient,  $R^2 = 0.9325$ , was obtained between the measured values of radon concentration in the winter period and the estimated annual average concentration. This result is useful for future research purposes, where it would be necessary to assess the annual average indoor radon concentration from the data based on one single seasonal radon indoor measurement. The later result suggested that the prediction from the average annual radon concentration is best done from winter measurements. Spring and autumn measurements should be used rather than summer measurements.

The geometric mean values of indoor radon concentration over municipalities of Skopje, measured in different season are presented on Figure 6. The results confirmed the known fact that the indoor radon concentration strongly varied with the season in the year. The highest radon concentration was observed in the winter, while the lowest concentration appeared in summer (LSD, p<0,05).

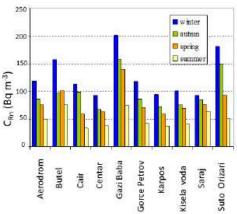


Figure 6. Geometric mean of indoor radon concentration in the municipalities of Skopje, measured in different season.

#### 4. CONCLUSION

- The annual mean indoor radon concentrations obtained for the studied area varies from 18 to 502 Bq m<sup>-8</sup>, which indicate that in some dwellings indoor radon concentration is not negligible. In general, significant difference between annual mean in different municipality were observed.
- The building characteristic: floor level, presence /absence of basement and type of heating significantly affecting a indoor radon concentration. The indoor radon concentration is generally higher in ground floor in houses without basement which use solid fuel type of heating
- Seasonal variation of indoor radon concentration is an important factor, were modeled by linear function.

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