Review

RESULTS OF INDOOR RADON MEASUREMENTS IN THE REPUBLIC OF MACEDONIA: – A REVIEW –

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Radon and its short lived decay products accumulated in indoor environment are the main source of public exposure to natural radiations. The health effects as well as a great number of natural and artificial factors affecting the radon accumulation in indoor environments are some of the motives for the scientific interest in radon issue. Following this global trend, many studies of indoor radon in the Balkan region, including the Republic of Macedonia have been conducted in the last decade. This paper is an overview of the published papers regarding indoor radon concentration measurements with nuclear track detectors in the Republic of Macedonia. It gives basic information about the spatial and temporal variability of indoor radon over the territory of the country, following by a description of the some factors which affect its variations. This review attempts: to organize available indoor radon results in order to show clear picture of the so far conducted surveys; to highlight the need for continuation of more extensive radon investigation in workplaces; to motivate the building professionals to create as much as possible mitigation methods for indoor radon reduction, to motivate the health professionals for epidemiological studies etc.

Key words: indoor radon; spatial variations; temporal variations; geology; building characteristics

INTRODUCTION

Radon (^{222}Rn) is a radioactive, noble gas produced by the decay of ^{226}Ra contained in all terrestrial materials. Both of them originate from ^{238}U decay chain. Radon decays with 3.8 days half-life by alpha particle emission followed by a chain of subsequent short lived decay products: ^{218}Po ($T_{1/2} =$ 3.05 min), ^{214}Pb ($T_{1/2} = 26.8$ min), ^{214}Bi ($T_{1/2} = 19.7$ min) and ^{214}Po ($T_{1/2} = 164$ μs).

Radon and its decay products accumulated in indoor environment are the main natural source of total public exposure to ionizing radiation (UNSCEAR, 2000). Some epidemiological studies have proven the association between the chronic exposure to high indoor radon concentrations (C_{Rn}) and the incidence of lung cancer (Darby et al., 2005). Considering serious health effects, the preparation and implementation of a national radon program (NRP), which main goal is the reduction of the radon risk,

becomes a primary task in most countries over the world. The NRP involves a complex organizational structure that includes components for radon monitoring, prevention and mitigation of increased C_{Rn} (WHO, 2009). Its preparation requires a multidisciplinary approach conditioned by the understanding of the radon processes in indoor environment.

The source of indoor radon is the radium present in soil and building materials (UNSCEAR, 2000). After its generation, radon emanates from solid grains and transported through porous media until its exhalation from the surface into atmosphere or into indoor environment. Diffusion and advection are the radon transport mechanisms driven by the concentrations and pressure gradients, respectively. In general radon dynamics is complex and depends on many factors resulting in the high variability of radon concentration in indoor air.

The factors that influence the C_{Rn} variation could be classified into three groups: geogenic

radon potential (GRP), building characteristics and building users lifestyle (habits). GRP is a factor that relates building underlying geology and C_{Rn}. The GRP is explained through the soil gas radon concentration and soil permeability. The soil gas radon concentration depends on the ^{226}Ra specific activity in the soil, which is proven to be in relation with geology. Permeability of the soil is another key geological factor that primarily affects the dynamics of radon in the soil gas as well as its emanation from the soil surface. The density of infiltrated radon flux from the soil, as well as accumulated radon concentration in the building, depends on the characteristics of the building. In the following, different habits of building inhabitants which relate to diverse levels of room ventilation (windows opening) is in function of the inhabitant's lifestyle, but also with meteorological conditions (among other: Yarmoshenko et al., 2016; Nikolopoulos et al., 2014).

The meteorological conditions also significantly affect the radon dynamics resulting in temporal variability of C_{Rn}. Just like the meteorological parameters, the indoor radon concentrations are subject to daily, weekly, monthly and seasonal variation (Kolarž et al., 2017; Nafezi et al., 2014; Ćurguz et al., 2013; Vaupotič, 2012; Vaupotič et al., 2008; Stojanovska et al., 2011). Because of that, the requirements are to present C_{Rn} in a building as annual C_{Rn}.

The awareness of the potential health problems that could be caused by the increased C_{Rn}, led to gradual growing of the investigations of indoor radon over last decades. As in the rest of the world, a large number of scientific works the Balkans, including the Republic of Macedonia, have been done. The papers that appeared in the literature are mainly related to radon sources, radon measurements and are focused as well on factors that affected radon variability (such as: Žunić et al., 2017a, 2017b, 2014, 2013, 2010, 2007; Ivanova et al., 2016; Vucković et al. 2016; Bochicchio et al., 2014; Vaupotič at al., 2013, 2008; Carpentieri et al., 2011).

This study summarized the results of indoor radon concentrations (C_{Rn}) in the Republic of Macedonia published in the literature. These results are based on radon measurements in 520 dwellings, 74 schools and 5 kindergartens. Discussion is oriented to radon spatial variations, the effect of buildings characteristics on radon variation, as well as to seasonal variations. The measured data is also compared with reported data for some countries in the Balkan region.

MATERIAL AND METHODS

Study area

The Republic of Macedonia is situated in the central part of the Balkan Peninsula (Southeastern Europe), covering an area of 25.713 km^2. Its geographical position is shown on the left maps in Figure 1. The territory is organized into 84 municipalities contained in eight statistical regions (separated with lines in Figure 1), with a total population of 2.022.547 (State Statistical Office, 2002).

Figure 1. Left two maps: Geographical position of the Republic of Macedonia. Right map presents four Geotectonical Zones and one area within the country coloured with different colours. The territory is organized in 8 statistical regions where the names are coded with capital letters: Skopje (SKO), Polog (POL), Southwest (SW), Pelagonia (PEL), Northeast (NE), East (East).

The area is described by complex geo-tectonic features of diverse relief and a complex geology. It is broadly divisible into four geotectonic units: Western Macedonian zone, Pelagonian massif, Vardar zone and Serbo-Macedonian massif and a separate Kratovsko-Zletovska volcanic rock area situated between the Vardar zone and the Serbo-Macedonian massif (five different colored in Figure 1). The entire territory has a transitional climate, from Mediterranean to continental.

Radon measurements

In general, the C_{Rn} measuring methods are active or passive. There is also a consequent application depending on the goals of the measurements. So, the active methods are used to make short term measurements and they are applied in cases when quickly respond to C_{Rn} is needed in some building or to examine radon dynamics for a certain relative-ly short period of time. The passive techniques are time integrated methods in which C_{Rn} is measured by nuclear track detectors, exposed over longer period usually in 3, 6 or 12 months. Detector response (calibrated in Bq/m^3) presents the average C_{Rn} for a certain period. The main advantage of this technique is the possibility to perform simultaneous C_{Rn} measurements in many buildings in a particular region.

According to the goals of the four radon surveys performed in Macedonia, all measurements were made using passive nuclear track detectors. The survey code, study area, type of environment with the number of buildings considered in the survey, floor level, detector commercial name, as well as the period of detector exposure for each survey separately are given in Table 1.

The principle of detection, as well as the results processing procedures of the measured concentrations, are explained in detail in the cited references in Table 1.

Table 1. Basic information for applied methodology in four C_{Rn} surveys in the Republic of Macedonia

Survey code	Study area	Type of environment (number of buildings)	Floor level	Detector commercial name	Detector exposure	Reference
S1	Whole country territory	Dwellings (437)	Ground, First Second	RSKS, RADUET	4 successive seasons (December 2008 to November 2009)	Stojanovska, 2011a Stojanovska, 2011b
S2	Five municipalities in Vardar zone	Schools (43)	Ground	TASTRAK RADUET	3 months in spring of 2012	Stojanovska, 2014
S3	Two municipalities in Kratovsko Zletovska area and one in Vardar zone	Dwellings (40), schools (31), kindergarten (5)	Ground	RSKS, Gamma 1	Annual (from June 2013 to May 2014)	Stojanovska, 2016a Stojanovska, 2016b
S4	Whole country territory	Dwellings (43)	Ground	RSKS	3 months in winter of 2013	Stojanovska, 2017

Design of surveys

The purpose of the first survey, marked by S1 in Table 1, was to established the basic map of the annual C_{Rn} distribution across the whole country territory (Stojanovska et al., 2011a). Seasonal variations of C_{Rn} were also studied (Stojanovska et al. 2011a). To that end, the detectors were deployed in the dwellings on the whole territory of the country. The number of detectors in each municipality was determined in dependence of the population density. The C_{Rn} was measured in 4 consecutive seasons, so the annual C_{Rn} was calculated as arithmetic mean of the 4 measurements.

In order to investigate the C_{Rn} variability in primary schools, the S2 survey was conducted in 5 municipalities in the geotectonical Vardar zone. The measurements were done in the spring of 2012 and the annual concentration was assessed using seasonal correction factors (Stojanovska et al., 2011b).

The third S3 survey, was conducted in 3 municipalities, one in the geotectonical Vardar zone and the other two in the Kratovo-Zletovska area. The detectors were deployed in the dwellings, primary schools and kindergartens and exposed for a period of one year.

The fourth research (S4) deals with the seasonal variability of C_{Rn} and the effects of

seasonal correction on the spatial variability of annual C_{Rn} and its uncertainty. The detectors were exposed during the winter period in 2013, in the dwellings throughout the whole territory of the country. The annual C_{Rn} was estimated according to the linear model developed in this study.

In all surveys and for each building, the measurements were made in one of the mostly occupied rooms: a living room or bedroom in the dwellings, classroom in the schools, and a playroom or bedroom in kindergartens.

RESULTS

The maximum value (Max), the arithmetic mean (AM), the standard deviation (SD), variation coefficient (CV = SD/AM), the geometric mean (GM) and geometric standard deviation (GSD) of the annual mean radon concentrations, as well as the number (percent) of buildings where indoor radon concentrations exceed national action level of 400 Bq/m³ for existing buildings, are given in Table 2.
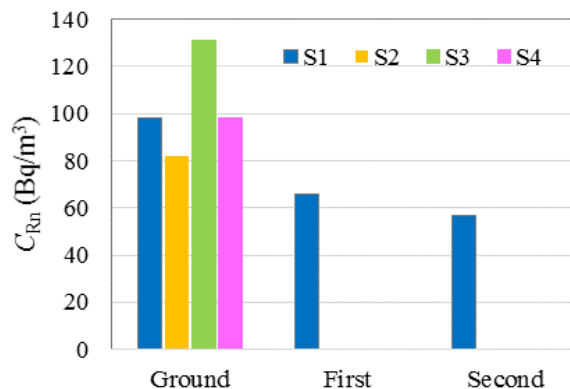
Table 2. Basic descriptive statisticc of annual radon concentrations measured in 4 radon surveys in the Republic of Macedonia

| Survey code | C_{Rn} | | | | | | N (%)>400 Bq/m³ |
	Max (Bq/m³)	AM (Bq/m³)	SD (Bq/m³)	CV (%)	GM (Bq/m³)	GSD	
S1	720	105	84	80	84	1.9	(1.8%)
S2	260	94	54	57	82	1.7	–
S3	990	186	178	95	131	2.34	7 (9%)
S4	460	120	85	69	98	1.9	1 (2%)

The mean values in the S1, S2, S3 and S4 studies expressed through AM and GM were in intervals from 94 Bq/m³ to 186 Bq/m³ and from 82 Bq/m³ to 131 Bq/m³, respectively. The C_{Rn} variations in each of them indicated by CV and GSD covered intervals: from 57% to 95% and from 1.7 to 2.34.

Building factors affecting radon variability

The GMs of annual C_{Rn} measured on the different floors in S1 survey in comparison with GM of annual C_{Rn} measured on ground floors in other three surveys are presented in Figure 2.

The GMs of C_{Rn} (with GSD in brackets) measured on ground, first and second floor in S1 were: 98 (1.9) Bq/m³, 66 (1.7) Bq/m³, 57 (1.6) Bq/m³, respectively. The GM values which represented S2, S3 and S4 surveys in Figure 2 are the same values given in the Table 2, respectively, since all measurements in that surveys were performed on the ground floors.

Figure 3 shows the GMs of annual C_{Rn} grouped according to the presence or absence of basement in buildings where measurements were done. The GMs of annual C_{Rn} (with GSD in brackets) in buildings with basement: 73 (1.8) Bq/m³, 83 (2.10) Bq/m³, 71 (1.65) Bq/m³, and without basement: 106 (1.8) Bq/m³, 161 (2.27) Bq/m³, 143 (1.7) Bq/m³ were obtained in S1, S3 and S4 surveys, respectively.

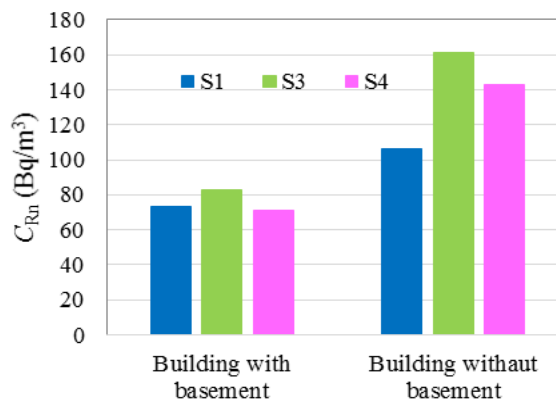**Figure 2**. GM of annual C_{Rn} measured in different building floors

Figure 3. GMs of annual C_{Rn} measured in buildings with and without floor
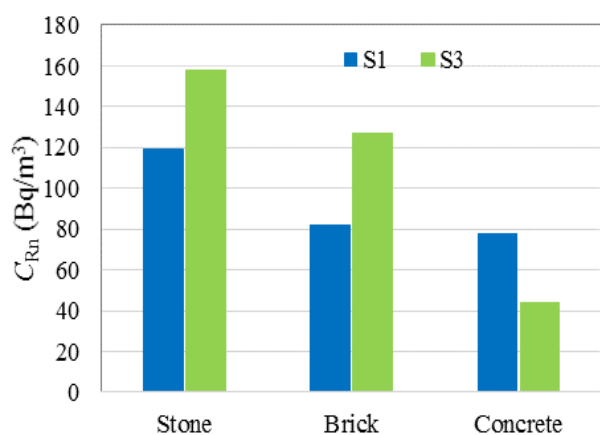
Figure 4. GMs of C_{Rn} measured in buildings with different dominant building materials

The effects of the type of dominant building materials on C_{Rn} variations were investigated in S1 and S3 surveys. Respectively for S1 and S3, the GM of C_{Rn} (with GSD in brackets) for buildings built out of stone: 119 (2.0) Bq/m^3 and 158 (2.41) Bq/m^3; brick 82 (1.9) Bq/m^3 and 127 (2.2) Bq/m^3; concrete 78 (1.8) Bq/m^3 and 44 (1.37) Bq/m^3 are presented in Figure 4.

Radon seasonal variation

The GMs of C_{Rn} (with GSD in brackets) measured in winter, spring summer and autumn were: 115 (2.02) Bq/m^3, 72 (1.97) Bq/m^3, 46 (1.95) Bq/m^3, 92 (2.02) Bq/m^3, respectively for S1 survey, while for S2 and S4 surveys these values were: 76 (1.7) Bq/m^3 and 114 (1.9) Bq/m^3 and corresponded for the measurements performed in spring and winter.

Comparison with literature

For comparison, the results from radon surveys performed in some European countries situated on the Balkan Peninsula are given in Table 3.

Table 3. Results from radon surveys performed in some countries on Balkan Peninsula

Country	Indoor (N)	Period of detector exposure (months)	C_{Rn} (Bq/m^3)			Reference
			Max	AM (SD)	GM (GSD)	
Serbia/Sokobanja	Dwellings (40)	12	189	55	43	Žunić et al. 2017
Serbia/Sokobanja municipality	Dwellings (43)	12	230	60 (44)	49 (1.8)	Mishra et al. 2014
Serbia/Niska Banja	Dwellings (65)		>6000	1163	529 (3.9)	Žunić et al. 2007
Serbia/Krusevac, Brus, Blace and Kursumlija	Dwellings (60)	3	358	82.3 (60.6)	65.6 (2.1)	Vucković et al. 2016
Serbia/ Southern part	Schools (207)	2 × 6	428	118 (78)	97 (1.9)	Žunić et al. 2013
Serbia /Kragujevac City	Kindergartens (14) Schools (28)	3	145	59.6 (25.6)	55.1 (1.18)	Stajić et al. 2015
Kosovo and Metohija/ South-Easter and Central part)	Dwellings (25)	2 × 6	378	163 (84)	143 (1.7)	Gulan et al. 2013
Kosovo and Metohija/ whole territory	Dwellings (48)	2 × 6	1016	122 (167)	71 (2.7)	Gulan et al. 2012
Republic of Srpska/ Banja Luka city	Schools (25) Schools (207)	12	549	128 (111)	99 (1.94)	Ćurguz et al. 2015
Bulgaria/Sofia city	Kindergartens (296)	3	1415	132 (118)	101 (2.08)	Ivanova et al. 2014
Bulgaria/ Sofia city, Sofia district, Plovdiv and Varna	Dwellings (373)	6	3560	158 (304)	99 (2.25)	Ivanova et al. 2013
Bulgaria/ Kremikovtsi municipality	Schools and kindergarten (9)	8	1761	694 (504)	542 (2.06)	Vuchkov et al. 2013
Greece/Xanthi prefecture	Schools (77)	8/10	958	231 (150)		Clouvas et al. 2009

DISCUSSION

Table 2 summarizes the basic statistics of the C_{Rn} measurements results obtained in the four surveys implemented in the Republic of Macedonia. The measured C_{Rn} values were observed to be normally distributed. The main conclusions from S1 were that C_{Rn} values have shown spatial variability throughout the country territory. The variabilities of C_{Rn} between country regions (Figure1), as well as within them, were significant. In general, they were mainly affected by regional and local geology features, as well as by the building characteristics. The maximum value of 720 Bq/m^3 reported for S1 survey was measured in Pelagonia statistical region (Stojanovska et al., 2011a;). In order to clarify this high value, a small measuring champagn in Kruševo (a small town in the Pelagonia statistical region) was conducted after S1 survey. The elevated values for C_{Rn} and natural radionuclides content in top soil were related to Amphibole-biotite granodiorite lithostratigraphy (Stojanovska et al. 2012).

Certain differences between results in the surveys given in the Table 2 are noticeable. They originated from the differences in each study design. For example, the S1 survey was conducted in dwellings distributed throughout the whole country territory, just like S4, but in the S4 measurements were organized only on the ground floor rooms. For these reasons, the C_{Rn} values in S4 were higher than in S1 survey. It is interesting to note that in these two studies the percentage of dwellings that exceeds the National action level of 400 Bq/m^3 is the same and it is 2% in both cases. Subsequently, the S2 survey was done on the ground floor in the Vardar zone, where from the previous knowledge elevated C_{Rn} in that area was not expected. The results confirmed the same, the obtained C_{Rn} results in S2 were lower than those corresponding to S1, also the maximum measured value was < 400 Bq/m^3. The higher C_{Rn} in Table 2 corresponds to S3 survey. In this case, the larger part of the investigated region lies down in Kratovska-Zletovska area, where bed rocks are from the volcanic origin. Detailed investigations of the C_{Rn} have been shown that the elevated C_{Rn} was in relation with tuff, tuffite of andesite and latite (Stojanovska et al., 2016). The existence of a connection between natural radioactivity and geology on the basis of broader research has been confirmed, not only due to indoor radon, but also to indoor thoron (Stojanovska et al. 2013) and ^{226}Ra (Bossew et al. 2013) in spatial variability analysis of surface soils.

The effect of the building floor on annual C_{Rn} variations can be seen in Figure 2. The GMs values of C_{Rn} measured in the ground floor in all surveys were higher than this in the first and second floors. On the other hand, the difference between the first and second floor concentrations is not significant (Stojanovska et al., 2011a;). Also the GMs values that correspond to the measurements in ground floors over the entire territory of the country (S1 and S4) are practically the same (Figure 2). Opposite to this, the results from S2 survey are lower and from S3 survey are higher than the GMs in S1 and S4 surveys. The GMs of the C_{Rn} measurements performed in buildings without basements are lower than in buildings with basements (Figure 3). Furthermore, the GMs of C_{Rn} in buildings with basements cover wider range than the GMs of C_{Rn} in buildings without basement. So, the spatial distribution of C_{Rn} in the buildings with basements is practically uniform, while in those without basements where the inhabitants are in direct contact with the ground the C_{Rn} varied considerably. These variations are associated with different influences of the geogenic radon over the different parts of the country.

From Figure 4, it can be concluded that the higher values of C_{Rn} refer to the stone buildings. On the same figure, the differences between the C_{Rn} measured in buildings made of brick and concrete are not noticeable for the S1 survey. On the contrary, the higher C_{Rn} in buildings built from bricks than these from concrete in the Kratovo-Zletovska area implicate the use of local building materials.

The fact that the global trend to improve energy efficiency in the buildings increase the C_{Rn} was confirmed in the S3 survey where the impact of new windows in buildings on C_{Rn} variations was studied. The analysis showed that the C_{Rn} in buildings with new windows was twice higher in comparison to C_{Rn} in buildings with wooden windows (GM = 110 (2.13) Bq/m^3). In the same survey, the environmental impact on C_{Rn} was examined, but it turned out that there was no difference between urban and rural areas (Stojanovska et al. 2016a).

That long term measurements of C_{Rn} in schools can be representative for a given region as well as for dwellings, even the different occupation time in schools was founded in the S3 survey. The results of annual C_{Rn} measurements in schools, dwellings and kindergartens were not different (Stojanovska et al., 2016a). There are also no differences between the results of C_{Rn} in schools when they are based on the annual measurements (12 months) and when they are in the period exempt from the summer holiday (9 months) (Stojanovska et al., 2016b).

The C_{Rn} seasonal variations are presented in Figure 5. The results clearly showed higher C_{Rn} for

the winter and autumn periods compared to spring and summer. The latter could be ascribed to the fact that during the colder months the buildings are heated, which creates a higher difference in pressure between the soil and the building. On the other hand, in order to save energy in cold periods, the inhabitants keep windows closed. The seasonal measurements in S1 survey were used to developed seasonal correction factors for annual C_{Rn} assessment due to C_{Rn} measurements in one season. The regional variability of the correction factor was confirmed only for the measurements in autumn (Stojanovska et al., 2011b). Applying the seasonal correction on measurements in winter season: the relative uncertainty budget of the annual C_{Rn} increased only for 3% as well as the regional variability over the country is not affected significantly (Stojanovska et al., 2017).
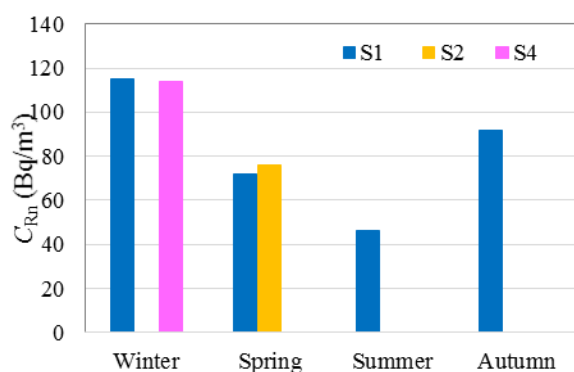

Figure 5. GM of C_{Rn} measured in different season

Finally we compared the obtained annual C_{Rn} in our research with that published in the literature. In general, the obtained GMs of C_{Rn} in all surveys (Table 2) were higher than the worldwide GM of 30 Bq/m³ reported in the UNSCER 2000 report. On the other hand, the both mean radon concentrations expressed with AM and GM, together with its deviations, are more or less typical as the radon levels reported for some other countries in the Balkan Peninsula (Table 3).

CONCLUSION

Spatial variations of C_{Rn} have been proven in all radon studies conducted in the Republic of Macedonia. In general, in the regions where bed rocks are from volcanic origin the C_{Rn} is higher in comparison with the other parts of the country. Also, the characteristics of the buildings have a significant effect on C_{Rn} variations. The highest concentrations were measured in buildings on the ground

floor without basements built with stone. When new windows are installed in such a building, the C_{Rn} increases additionally. The results show that in the few percents of buildings in Macedonia the C_{Rn} exceeds the permissible level and they have to be remediated. Further research in this field should be include health and building professionals in order to create an effective radon protection program.

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РЕЗУЛАТИ ОД МЕРЕЊАТА НА РАДОН ВО ЗАТВОРЕН ПРОСТОР НИЗ РЕПУБЛИКА МАКЕДОНИЈА: ПРЕГЛЕД

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Акумулираниот радон и неговите продукти со кусо време на распаѓање во затворен простор се главен природен извор на озрачување на населението. Ефектите врз здравјето, како и бројните природни и вештачки фактори кои влијаат врз акумулацијата на радон во затворен простор, се еден од мотивите за научниот интерес за радонот. Следејќи го овој глобален тренд на Балконот, вклучувајќи ја и Република Македонија, во последната деценија беа спроведени многу истражувања на радонот во затворен простор. Овој труд дава преглед на веќе објавените резултати од спроведените мерења на концентрациите на радон во простории низ Република Македонија користејќи нуклеарни трагови детектори. Тој дава основни информации за просторната и временската варијабилност на радонот во затворен простор низ територијата на земјата, проследено со опис на ефектите од некои фактори кои влијаат врз тие варијации. Намерата на прегледот е: да ги организира и да ги искоментира достапните резултати за радонот во затворен простор, со цел да се даде јасна слика за досега спроведените истражувања, да се истакне потребата од продолжување на пообемно истражување на радонот во просториите на работните места, мотивирање на градежните професионалци за креирање колку што е можно повеќе мерки за ублажување и превенција од зголемини концентрации на радон во затворен простор, мотивирање на здравствените работници за епидемиолошки студии итн.

Клучни зборови: радон во затворен простор; просторни варијации; временски варијации; геологија; карактеристики на зграда