THE INDOOR RADON AND THORON CONCENTRATIONS IN SCHOOLS OF SKOPJE (REPUBLIC OF NORTH MACEDONIA) AND BANJA LUKA (REPUBLIC OF SRPSKA) CITIES MEASURED BY RADUET DETECTORS

Zdenka Stojanovska1,*, Zoran Ćurguz2, Predrag Kolarž3, Zora S. Žunić4, Ivan Boev5, Blazo Boev5

1 Faculty of Medical Sciences, Goce Delčev University Stip, Krste Misirkov No.10-A P.O. Box 201, Republic of Macedonia
2 Faculty of Transport and Traffic Engineering, University of East Sarajevo, Vojvode Mišića 52, 74 000 Doboj, Republic of Srpska
3 Institute of Physics, University of Belgrade, Serbia,
4 Institute of Physics Belgrade, Pregrevica 118, 11080 Zemun, Serbia
5 Faculty of Natural and Technical Sciences, Goce Delcev University, Stip, Krste Misirkov10-A, P.O 201, Republic of Macedonia

Abstract: Radon (222Rn) and thoron (220Rn) are natural radioactive gases, generated in the terrestrial materials. They are the main sources of public exposure to ionising radiation in any of indoor environment worldwide. Differences in half-lives of 222Rn (T_{1/2} = 3.8 d) and 220Rn (T_{1/2} = 55.6 s) lead to its different indoor behavior. Several studies of indoor 222Rn and 220Rn in Northern Macedonia have been performed, starting with measurements in dwellings in 2008 and continuing with measurements in schools during 2012. The surveys in the Republic of Srpska began later (in 2011) with the simultaneous 222Rn and 220Rn measurements in the dwellings and schools of Banja Luka cities. This paper, as a result of our cooperation, summarizes the results and general conclusions obtained from 222Rn and 220Rn measurements in schools of capitals. In both cities, the measurements were made using Raduet - nuclear track detectors; deployed at distances: >0.5m (Skopje) and 0.2m (Banja Luka); and exposed in a period: March 2012 - May 2012 (Skopje) and April 2011 - May 2012 (Banja Luka). Results for 222Rn and 220Rn concentrations in both cities have a log-normal distribution. The 222Rn geometric mean value of 71 Bq/m³ in Skopje is higher than in Banja Luka city (GM = 50 Bq/m³). Among different radon potential in the cities, this difference could be related to the different exposure time of detectors. Furthermore, the dispersion of the 222Rn results in each city expressed through geometric standard deviation is relatively low: GSD = 2.13 (Skopje) and GSD = 2.11 (Banja Luka) indicating relatively homogeneous data sets. The 220Rn concentrations in Banja Luka city (GM = 51 Bq/m³) were higher than in Skopje (GM = 11 Bq/m³). It is obvious that in the case of 220Rn, the exposure period did not play a significant role. One of the reasons for this difference could be the position of the detectors as well as the different building materials in the schools. On the contrary, the dispersion of the 220Rn results in Skopje (GSD = 3.38) was greater than in Banja Luka (GSD = 2.07).

Keywords: radon, thoron, gas, school.

1. INTRODUCTION

Radon (222Rn) and thoron (220Rn) are radioactive gases from the respective natural series of 238U and 232Th, which are present in the terrestrial materials from the earth formation to the present day. The existing trace concentrations of 238U and 232Th, and their corresponding decay products 226Ra and 222Ra in building materials and in the underlying rocks and soil are sources of radon and a thoron in a building. The generated gases move through enclosed spaces of the material, emanate from its surface and enter into other environments where they can be accumulated. The dynamics are complex and driven

*Corresponding author: zdenka.stojanovska@ugd.edu.mk
mainly by the processes of advection and diffusion. Amount of the accumulated radon or thoron indoors concentration, depends on many factors. In general, they are related to the concentration of the radionuclides in the terrestrial material together with its porosity (radon potential), building characteristics and mode of its use. Additionally, the advection process is in a direct function of the outdoor-indoor temperature gradient, after all, radon and thoron concentration variations strongly depend on the meteorological parameters (temperature, pressure, wind, etc.).

Radon has a relatively long half-life ($T_{1/2} = 3.8$ d), therefore the gas created in underlying soil in high depths can enter and accumulate in a building before it decayed. Contrary, due to short half-life ($T_{1/2} = 55.6$ s), thoron can travel only short distances before it decayed. The main consequences of this are the differences in the effect of the factors on the accumulation of these two gases in indoor environments. In general, according to a large number of studies [1-3], it can be said that the main source of radon is the underlying soil (rock) and to a lesser extent the building materials, while in the case of thoron: the building materials are the dominant source in a building. Also, the essential difference between these two gases is that the indoor radon concentrations are homogeneously distributed, unlike the indoor thoron, whose concentrations are at the maximum on the wall and decrease at distance from it. Moreover consequence of the differences in radon and thoron half-lives, are discrepancy in their temporal variations caused by the meteorological parameters. For example, they are more pronounced for radon than for thoron [4-9].

Based on a large number of studies, it has been shown that indoor radon and thoron, are dominant (over all) radioactive sources to public exposure [9-10]. The harmful health effects of chronic exposure to them, have been confirmed [11], so the indoor radon and indoor thoron are very important radiation protection issue in every country. Following world trends, numerous campaigns to measure indoor radon and thoron in the countries of the Balkan, have been performed in the last decade. Thus, many papers have been published, some of which are cited in this paper’s references [12-22]. In most of them radon concentrations are measured while studies of indoor thoron concentrations are limited. Depending on the type of indoor environment, the studies were conducted mainly in dwellings, schools (kindergartens) or combined. In Republic of North Macedonia, the first indor radon and thoron measurements were made in dwellings in 2008 [23] and only later (2012) in schools and kindergartens [24-26]. In Republika Srpska, an entity in Bosnia and Herzegovina, the first systematic survey indoor radon and thoron was launched in 2011, simultaneously measuring both dwellings and schools [27-28]. Most of the results of these studies are already published.

The main purpose of this paper is to show the extracted results of radon and toron concentration measurements, previously performed as a part of the more complex survey. In this study, we considered, only measured concentrations with the same type of detector, in the schools of the capital cities Skopje and Banja Luka. The geographical positions of the two republics in Europe, as well as the position of the cities, are shown in the Figure 1.
2. MATERIAL AND METHODS

The indoor radon and thoron concentrations were measured in the schools of Skopje and Banja Luka using a nuclear track detectors, with Raduet commercial name, manufactured in Radosys, Hungary (Figure 2). The operating principle of Raduet was explained precisely in our previous studies [6, 28].

In Skopje, the detectors were installed in a classroom on the ground floor at a distance of 0.5 m from any wall surface and exposed in a period of three months, from March to May 2012. In Banja Luka, they were deployed in a shorter distance of 0.2 m from the wall surfaces and exposed for one year, from April 2011 to May 2012. Types of the rooms, the distances to the wall surfaces along with the detectors period of exposure in each of the surveys are specified in Table 1.

![Raduet nuclear track detector produced in Radosys, Hungary](image)

**Figure 2.** Raduet nuclear track detector produced in Radosys, Hungary

| City          | Type of room           | Distance from wall surface | Period of detectors exposure          |
|---------------|------------------------|----------------------------|---------------------------------------|
| Skopje        | Classroom              | >0.5m                      | March-May, 2012                       |
| Banja Luka    | Assembly hall or hallway | 0.2 m                     | April, 2011-May, 2012                 |

3. RESULTS

The histograms of the measured indoor radon ($^{222}\text{Rn}$) and thoron ($^{220}\text{Rn}$) concentrations in both cities are shown in Figure 3. The best fit for both gases data sets was the log-normal function whose parameters are also present on the graphs in Figure 3.

![Histogram of 222Rn](image)

**Figure 3.** Histograms fitted with a log-normal function of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ concentrations measured in schools of Skopje and Banja Luka
The number of schools under observation (N), number of schools with measured concentration below detection limit (N_{\text{MDA}}), arithmetic mean (AM), standard deviation (SD), geometric mean (GM) and geometric standard deviation (GSD) of indoor radon (\(^{222}\text{Rn}\)) and thoron (\(^{220}\text{Rn}\)) concentration measured in the schools of Skopje and Banja Luka cities are given in Table 2. In schools of Skopje, radon values range from 9 to 379 Bq/m\(^3\) and those of thoron range from 1 to 80 Bq/m\(^3\). The ranges in schools of Banja Luka are 25-341 Bq/m\(^3\) for radon and 7-198 Bq/m\(^3\) for thoron. In 12 schools of Skopje, thoron concentration was below the detection limit of 1 Bq/m\(^3\). Except in two schools, the radon concentration in Skopje were higher in comparison to measured thoron concentrations. In Banjaluka, the thoron concentrations were higher than radon concentrations in most cases. The intervals of \(^{220}\text{Rn}/^{222}\text{Rn}\) ratio in the schools considered in the present study were 0.01 - 4.72 for Skopje and 0.14-3.80 for Banja Luka (Figure 4) with geometric mean values of 0.19 and 1.00, respectively.

### Table 2. Basic statistic of indoor radon and thoron concentrations considered in this study

|          | N  | N_{\text{MDA}} | AM Bq/m\(^3\) | SD Bq/m\(^3\) | GM Bq/m\(^3\) | GSD  |
|----------|----|----------------|---------------|---------------|---------------|------|
| \(^{222}\text{Rn}\) | 58 | 0              | 85            | 81            | 61            | 2.15 |
| \(^{222}\text{Rn}\) Skopje | 33 | 0              | 94            | 78            | 71            | 2.13 |
| \(^{222}\text{Rn}\) Banja Luka | 25 | 0              | 72            | 85            | 50            | 2.11 |
| \(^{220}\text{Rn}\) | 58 | 12             | 42            | 39            | 25            | 3.42 |
| \(^{220}\text{Rn}\) Skopje | 33 | 12             | 18            | 18            | 11            | 3.38 |
| \(^{220}\text{Rn}\) Banja Luka | 25 | 0              | 63            | 40            | 51            | 2.07 |

![Figure 4. The \(^{220}\text{Rn}/^{222}\text{Rn}\) ratio in schools of Skopje and Banja Luka](image)

4. DISCUSSION AND CONCLUSION

This study presents the results for indoor radon (\(^{222}\text{Rn}\)) and thoron (\(^{220}\text{Rn}\)) concentrations measured in schools of Skopje and Banja Luka cities with Raduet detectors exposed in periods of 3 and 12 months, respectively (Table 1). Analysis of the distribution of radon and thoron concentration showed that the best fit of the data was log-normal function. Visually (from Figure 3) and further by testing, the differences between log-normal functions of the Skopje and Banja Luka data sets, was confirmed for radon as well for thoron. The reason for this may be the different contributions of the factors which are affecting radon and thoron concentrations variation in the cities.

The arithmetic and geometric mean values of radon concentrations measured in Skopje are higher than the mean concentrations obtained for Banja Luka (Table 2). We assumed that, among the different radon potentials, the different types of rooms where measurements were performed as well the different periods of detectors exposure, could be a reason for this. But, lower radon concentrations in the hall in comparison to the classroom in Skopje and assembly halls in Banja Luka schools, were not confirmed, so the assumptions about differences in exposure period and radon potential remain. In Banja Luka, the radon concentrations measured by Raduet (this study) in one room are lower than those reported previously, where the radon for each school was presented as a
mean value of measured results, with two different types of detectors in at least two different types of rooms. Furthermore, the geometric standard deviation values (GSD) obtained for radon data from Skopje and Banja Luka (Table 2), are relatively low which indicate relatively homogeneous factors effects on radon variation in each city.

Contrary to radon, the thoron concentrations measured in schools of Banja Luka city are higher in comparison to those of Skopje city (Table 2). It is obvious that in the case of thoron, a period of the detector exposure does not play a significant role. Some of the reasons for this difference could be the position of the detectors as well as the different building materials in the schools. The dispersion of the results in Skopje is greater than the result measured in Banja Luka.

For comparison, the geometric mean concentrations of radon in Banja Luka schools (GM = 50 Bq/m³) are lower than those reported by studies conducted in schools: in Skopje (GM = 71 Bq/m³), Eastern part of North Macedonia (GM = 96 Bq/m³) [25], (GM = 87 Bq/m³) [26], Southern Serbia (GM = 97 Bq/m³) [29], Osijek, Croatia (GM = 70.6 Bq/m³) [30], as well as lower than those published for kindergartens in Sofia, Bulgaria (GM = 101 Bq/m³) [31] and Kremikovtzi Municipality, Bulgaria (GM = 542 Bq/m³) [32]. The GM = 51 Bq/m³ for thoron measured in Banja Luka schools is not only higher than the GM = 11 Bq/m³ value obtained from measurements in Skopje, but also higher than the usual published values in the literature.

5. REFERENCES

[1] K. Ivanova, Z. Stojanovska, M. Tsenova, B. Kunovska, Building-specific factors affecting indoor radon concentration variations in different regions in Bulgaria, Air Quality, Atmosphere & Health, Vol. 10-9 (2017) 1151–1161.

[2] F. Bochicchio, Z. S. Žunić, C. Carpentieri, et al., Radon in indoor air of primary schools: a systematic survey to evaluate factors affecting radon concentration levels and their variability, Indoor Air, Vol. 24 (2014) 315–326.

[3] P. Bossew, Z.S. Žunić, Z. Stojanovska, et al., Geographical distribution of the mean radon concentrations in primary schools of Southern Serbia - application of geostatistical methods, J Environ Radioact, Vol. 127 (2013) 141–148.

[4] K. Ivanova, Z. Stojanovska, Modelling of the temporal indoor radon variation in Bulgaria, Radiat Environ Biophys, Vol. 58–3 (2019) 337–344.

[5] Z. Stojanovska, J. Janusiski, P. Bossew, et al., Seasonal indoor radon concentration in FYR of Macedonia, Radiat Meas., Vol. 46–67 (2011) 602–610.

[6] Z. Stojanovska, P. Bossew, S. Tokonami, et al., National survey of indoor thoron concentration in FYR of Macedonia (continental Europe - Balkan region), Radiat Meas., Vol. 49–1 (2013) 57–66.

[7] Z. Stojanovska, B. Boev, Z.S. Žunić, et al., Results of radon CR-39 detectors exposed in schools due two different long-term periods, Nukleonika, Vol. 61–3 (2016) 385–389.

[8] Z. Stojanovska, K. Ivanova, P. Bossew, et al., Prediction of long-term indoor radon concentration based on short-term measurements, Nucl Technol Radiat, Vol. 32–1 (2017) 77–84.

[9] United Nations Scientific Committee, Effects of Atomic Radiation Effects of ionizing radiation. Report to the General Assembly with Scientific Annexes, Annex B, NY: UN, 2000.

[10] United Nations Scientific Committee, Effects of Atomic Radiation Effects of ionizing radiation. Report to the General Assembly with Scientific Annexes, Annex B, NY: UN, 2008.

[11] S. Darby, D. Hill, A. Auvinen, et al., Radon in homes and risk of lung cancer: Collaborative analysis of individual data from 13 European case-control studies, (2005) BMJ 330.7485 223–0

[12] Z. S. Žunić, Z. Stojanovska, B. Boev, et al., Sjenica, a newly identified radon priority area in Serbia, and radon data correlated with geological parameters using the multiple linear regression model, Carpathian Journal of Earth and Environmental Sciences, Vol. 14–1 (2019) 235–244.

[13] Z. S. Žunić, R. Mishra, I. Čeliković, et al., Effective Doses Estimated from the Results of Direct Radon and Thoron Progeny Sensors (DRPS/DTPS) Exposed in selected Region of Balkans, Radiation Protection Dosimetry, Vol. 185–3 (2019) 387–390.

[14] K. Ivanova, Z. Stojanovska, B. Kunovska, et al., Analysis of the spatial variation of indoor radon concentrations (national survey in Bulgaria), Environmental Science and Pollution Research, Vol. 26–7 (2019) 6971–6979.

[15] Z.S. Žunić, P. Bossew, F. Bochicchio, et al., The relation between radon in schools and in dwellings: A case study in a rural region of Southern Serbia, J Environ Radioact, Vol. 167–1 (2017) 188–200.

[16] Z.S. Žunić, Z. Stojanovska, N. Veselinovic, et al., Indoor radon, thoron and their progeny concentrations in high thoron rural Serbia environments, Radiation Protection Dosimetry, Vol. 177–12 (2017) 36–39.

[17] P. Kolarž, J. Vaupotič, I. Kobal, et al.,
Thoron, radon and air ions spatial distribution in indoor air, J Environ Radioact, Vol. 173 (2017) 70–74.

[18] Z.S. Žunić, P. Ujić, L. Nad, et al., High variability of indoor radon concentrations in uraniferous bedrock areas in the Balkan region, Appl radiat Isotop, Vol. 94 (2014) 328–337.

[19] R. Mishra R. Z. S. Žunić, G. Venoso, et al., An evaluation of thoron (and radon) equilibrium factor close to walls based on long-term measurements in dwellings, Radiat Prot Dosimetry, Vol. 160–1–3 (2014) 164–168.

[20] L. Gulan, F. Bochicchio, C. Carpentieri, et al., High annual radon concentration in dwellings and natural radioactivity content in nearby soil in some rural areas of Kosovo and Metohija (Balkan region), Nucl Technol Radiat, Vol. 28–1 (2013) 60–67.

[21] K. Ivanova, Z. Stojanovska, V. Badulin, et al., Pilot survey of indoor radon in the dwellings of Bulgaria, Radiat Prot Dosimetry, Vol. 157–4 (2013) 594–599.

[22] L. Gulan, G. Milic, P. Bossew, et al., Field experience on indoor radon, thoron and their progenies with solid-state detectors in a survey of Kosovo and Metohija (Balkan region), Radiat Prot Dosimetry, Vol. 152–(1–3) (2012) 189–197.

[23] Z. Stojanovska, J. Januseski, B. Boev, et al., Indoor exposure of population to radon in the FYR of Macedonia, Radiat Prot Dosimetry, Vol. 148–2 (2011) 162–167.

[24] Z. Stojanovska, Z. S. Žunić, P. Bossew, et al., Results from time integrated measurements of indoor radon, thoron and their decay product concentrations in schools in the Republic of Macedonia, Radiat Prot Dosimetry, Vol. 162–1–2 (2014) 152–156.

[25] Z. Stojanovska, B. Voev B. Z. S. Žunić, et al., Variation of indoor radon concentration and ambient dose equivalent rate in different outdoor and indoor environments. Radiat Environ Biophys, Vol. 55–2 (2016) 171–183.

[26] Z. Stojanovska, B. Voev, Z. S. Žunić, et al., Factors Affecting Indoor Radon Variations: A Case Study in Schools of Eastern Macedonia, Rom J Phys, Vol. 64 (2019) 801.

[27] Z. Ćurguz, Z. S. Žunić, Tollefsen, et al., Active and passive radon concentration measurements and first-step mapping in schools of Banja Luka, Republic of Srpska, Rom J Phys, Vol. 58–S (2013) 90–98.

[28] Z. Ćurguz, Z. Stojanovska, Z.S. Žunić, et al., Long-term measurements of radon, thoron and their airborne progeny in 25 schools in Republic of Srpska, J Environ Radioact, Vol. 148 (2015) 163–169.

[29] Z. S. Žunić, C. Carpentieri, Z. Stojanovska, et al., Some results of a radon survey in 207 Serbian schools. Rom J Phys, Vol. 58–S (2013) 320–327.

[30] J. Planinić, G. Šmit, Z. Faj, et al., Radon in schools and dwellings of Osijek, Journal of Radioanalytical and Nuclear Chemistry, Vol. 191–1 (1995) 45–51.

[31] K. Ivanova, Z. Stojanovska, M. Tsenova, et al., Measurement of indoor radon concentration in kindergartens in Sofia, Bulgaria, Radiat Prot Dosimetry, Vol. 162–1–2 (2014) 163–166.

[32] Vuchkov D. K. Ivanova, Z. Stojanovska, et al., Radon measurement in schools and kindergartens (Kremikovtsi Municipality, Bulgaria), Rom J Phys, Vol. 58–S (2013) 328–335.
2012. (Бања Лука). Резултати концентрација 222Rn и 220Rn у оба града имају log-нормалну дистрибуцију. Средња геометријска вредност 222Rn од 71 Bq/m^3 у Скопљу је већа него у Бањој Luци (ГС = 50 Bq/m^3). Између разлике у потенцијалу радона који утичу на варијације 222Rn, та се разлика може повезати са различитим временима излагања детектора. Даље, дисперзија резултата 222Rn у сваком граду изражена геометријском стандардном девијацијом је релативно мала: ГСД = 2,13 (Скопље) и ГСД = 2,11 (Бања Лука). Иако су две геометријске вредности разликоване 50 и 51 Bq/m^3, резултати су већи у Бањој Луци. Очигледно је да у случају 220Rn, период изложености није играо значајну улогу. Један од разлога ове разлике могу бити положај детектора као и различити грађевински материјали у школама. Супротно томе, дисперзија резултата 220Rn у Скопљу (ГСД = 3,38) била је већа него у Бањој Луци (ГСД = 2,07).

Кључне речи: радон, торон, гас, школа.

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