The problem with indoor radon and solution in the use of geothermal water N. Chobanova^{1*}, B. Kunovska^{1*}, D. Djunakova¹, J. Djounova¹, A. Angelova¹, Z. Stojanovska², K. Ivanova¹

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Bulgaria is rich in geothermal water. The radon level in spas buildings depends on soil underneath, thermal water and the ventilation conditions. The aim of the paper is to analyses influence of ventilation system on indoor radon in air in the spa buildings. Direct measurements of radon in the air were performed in treatment rooms (treatment with thermal water and without) and mineral pools. A statistically significant difference was found between the indoor radon levels in treatment rooms using water and those without water, located in branches with high radon concentration (MW, p = 0.032). The spectra of the rooms for 24 hours were considered for analysing the indoor radon daily variations. The radon arithmetic mean value in rooms with operating ventilation system was 148 Bq /m³, and in those without - 756 Bq /m³ and the statistically difference is significant (MW, p < 0.0001). Further the radon levels decrease 1.7 to 3.3 times when the ventilation works. To improve air quality in Rehabilitation Centres, the ventilation system must be in good function. The mineral water is also a source of radon and varies according to its degree of use. The effective dose of a visitor who performed thermal procedures was also evaluated.

Keywords: Indoor radon concentration, geothermal water, ventilation system, spas, direct measurement, spectra

INTRODUCTION

Bulgaria is a country that is relatively rich in geothermal water, the temperature of which is in the range 25°C-100°C. The mostly (>95%) of natural mineral springs are concentrated in the southern part of Bulgarian territory. Generally, there are more than 170 geothermal fields in Bulgaria and numerous manifestations have been discovered and delineated (~ 1000 natural mineral springs and 2000 boreholes) [1, 2]. According to the Water Act, 102 of all hydrothermal fields in Bulgaria are specified as exclusive state property. The rest are municipal property [3].

Thermal waters can be used for electricity production (at temperatures > 150° C) or at lower temperatures for direct application heating of buildings and greenhouses, bottling of mineral water, etc. Traditionally, their greatest application is in the field of balneology. Most of them are very popular with the population in the area of prevention, medical therapy and rehabilitation as well as tourism and recreation [1].

The major factors that contribute to the geothermal development in Bulgaria are: long tradition, favourable climate, appropriate

thermal water composition and a developed spa system [1].

In spa environment radon and its decay products have been considered as the main ionizing radiation exposure for both users and workers therefore the radon in spas should be monitored [4]. Radon is recognized as a leading cause of lung cancer incidence, excluding tobacco [5, 6]. The risk of radon exposure is associated with mostly high radon concentrations in confined environments and the subsequent inhalation. Radon is formed from radium, dissolve in groundwater, which is contact with soil and rocks containing radium. Its high degree of mobility allows it to be transported to the surface and then diffuse into the atmosphere and buildings. Geothermal spas are one of the common confined places where radon may be present in higher concentrations due to aeration, small ventilation or constructive details [7, 8, 9, 10]. Such places must be subjected to special investigation, taking into account that radon level in spas buildings depends on both two potential factors (soil underneath and thermal water) and the ventilation conditions of various premises.

The European Directive 2013/59/EURATOM defined the reference level for the annual average activity radon concentration in houses, public buildings and workplaces should not be higher than 300 Bq.m³

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[11]. The requirements have been harmonized into Bulgarian national legislation [12].

The aim of the article is to analyse the influence of water as an additional source of radon in the air and daily variation of indoor radon in order to assess the effect of the ventilation system, as a possible solution to reduce the exposure of the population.

MATERIALS AND METHODS

Area of study

To achieve the purpose, the variation of indoor radon concentration in 11 branches of the Specialized Hospitals for Rehabilitation National Complex (85% of the national branches with thermal spas in Bulgaria), located on the territory of Bulgaria have been studied. These sites use geothermal water for treatment and that's why we agreed to call them spa. 36 treatment premises located in 11 branches were surveyed. The spa buildings are located in 9 districts of Bulgaria - one in Sofia district, one in Plovdiv district and the other 7 are in western part of Bulgaria, in the districts of Blagoevgrad, Kyustendil, Pazardzhik, Stara Zagora, Smolyan, Montana and Veliko Tarnovo, respectively. Fig.1 shows the location of the studied branches.



Fig.1. Location of the studied Bulgarian spas (1-Narechenski Bani, 2-Momin Prohod, 3-Hisarya, 4-Pavel Banya, 5-Varshets, 6-Bankya, 7-Kyustendil, 8-Sandanski, 9-Velingrad, 10-Banite,11-Ovcha Mogila)

The spas are open all year round. Many patients are treated in these spa complexes, which are recommended for respiratory diseases, dermatology, rheumatology, neurological diseases etc. They apply different services and treatment techniques as baths, jets, circular showers, inhalations, sprays, sauna, underwater massage and manual massage. The treatment rooms are supplied with thermal water which contains specific amounts of dissolved substances and together with the high temperature turn out beneficial health effects. Spring water is pumped from its source through a mechanical drainage system located a few meters outside the spa.

The variations of radon in the air of the studied premises have a great variability and can be controlled by many factors (ventilation, number of patients using the equipment, humidity, etc.). The ventilation in the treatment rooms, apart from the naturally is carried out through built-in ventilation system. a Information about the ventilation system in the premises was collected via specially prepared questionnaire and analysis of the influence of the factor was made too. There are working general or partial ventilation systems in 5 branches. In other six buildings there are no ventilation systems. In most branches the ventilation system works 4 hours a day, and in only one of them is less. A total of 36 measurements were performed in treatment rooms (treatment with thermal water and without) and mineral pools. 18 of the measured rooms are located on the ground floor, eight are in the basement, nine - on the first floor and only one on the second floor.

Measurement methods

Radon concentrations in the air of the spa complexes were measured by Alpha Guard PQ2000 radon monitor (Genitron Instruments, Germany), detector AlphaE (Saphymo GmbH, Germany) and TERA (TSR3D) system (Tesla, Czech Republic). The equipment also registered air temperature (-20° C to $+60^{\circ}$ C), humidity (10% to 90%) and energy spectrum. Measuring system Alpha Guard PQ2000 uses a principle of ionization chamber (contaminated the background of 40 Bq.m³); radon enters the detection area (through a Rn progeny preventing filter), where it is measured by α -spectrometric technique. The device is not affected by high humidity and vibration. Its range has a linear dependence from 2 Bq.m⁻³ to 2,000,000 Bq.m³. Data analysis, processing, and storage were performed using the DataEXPERT software. With this equipment radon measurement were performed in the spa centres for 24 hours in the period from February to June 2019 (12 measurements).

AlphaE is an active measurement device for detecting and recording the radon concentration. Based on the diffusion principle with silicon detector the radon gas diffuses through the entry holes of the case into the inner of the diffusion chamber. As the holes are entirely covered by a Gore-Tex membrane only the radon gas can enter the chamber while the decay products, are retained by the filter.

With TERA (TSR3D) system it is possible to continuously monitor the radon concentration in the range of 20 Bq.m³ to 1 MBq.m⁻³, with possibilities for measuring the instantaneous volume activities of radon: short-term (average hourly) and long-term (daily average). The measurement uncertainty is as follows <13% at 300 Bq.m³ for 1 hour; <3% at 300 Bq.m³ in 24 hours. The 24 hours' radon spectra were measured from October 2019 to February 2020 with newly received equipment, supported by the Bulgarian National Science Fund. Direct measurements and monitoring of the radon concentration over time allow clarification of the possible reasons for the dynamics of radon, as well as the formulation of specific conclusions about its distribution in time and space of the measured premises.

During these measurements, the devices Alpha Guard PQ2000 and AlphaE was operated in the 10 minutes' data sampling cycle while TERA (TSR3D) system for 1 hour. Direct radon measurements were performed in treatment rooms and mineral pools, which are located in ground floor, basement and first floor.

Information from questionnaire

Through a purposive questionnaire we received information about the type of building construction, year of construction, presence of mechanical ventilation systems, presence of elevators, does ventilation system works, done any energy saving measures, the types of windows etc. Such a questionnaire was completed by an employee in each spa branch.

Statistical analysis

To perform statistical analysis was used IBM SPSS Statistics, v. 23. The measurements are grouped as follows: types of premises (with water and without water) and premises with ventilation system (working and no working), to assess the radon variations. Descriptive analysis, parametric and non-parametric tests are applied for data processing. A parametric test of Kolmogorov-Smirnov was used to check the normality of the distribution of radon concentration in air and a non-parametric Mann-Whitney test was applied to compare two independent groups. Level of significance is p<0.05.

RESULTS AND DISCUSSION

The descriptive statistic of direct measurements of radon concentration in 36 spa premises is presented in Table 1. The radon concentrations are not following a log normal distribution (p<0.05). The obtained arithmetic mean and standard deviation have been found to be 604 Bq.m³ and 1020 respectively.

Table 1. Descriptive statistic of radonconcentrations measured in 36 premises

Premises	AM CRn	Min CRn	Max CRn	CV
	Bq.m ³	Bq.m ³	Bq.m ³	%
36	604	16	4455	169

The coefficient of variation is 169%, which shows large difference between the а concentrations of radon in the studied premises. This percentage is so high due to the large range of radon concentration from 16 Bq.m⁻³ to 4455 Bq.m⁻³. The large deviation of radon levels is due to different factors such as: location and construction of the building, the presence of a working ventilation system, the composition of the mineral water in the individual spa buildings.

We analysis two groups of premises: rooms using water and those who do not use water (Fig.2). The indoor radon is higher in rooms with water (AM=634 Bq.m³), than in rooms without water (AM=355 Bq.m³), indicating that the use of geothermal water for treatment increases radon levels in the air.

To explore the hypothesis if the thermal water affected the variations of radon in treatment rooms the measurements in two branches (Narechenski Bani and Momin Prohod) with high levels of radon are considered for detail analysis. A statistical test to confirm the hypothesis that the used mineral water is an additional source of radon in air of the studied premises was performed. The values of indoor radon concentration differ significantly between the studied premises - with water and without water (MW, p = 0.032). Thermal water is a radon source, which varies according to its degree of use in the treatment rooms. The analysis of the spectra indicates that usually the indoors Rn values vary depending on the work activities during the day. Our results are comparable with the values reported for spas in other regions worldwide [10, 13].



Fig.2. Distribution of radon concentration depending on the use or non-use of water in the treatment premises

In order to evaluate the efficiency of the ventilation system, the results of Narechenski Bani and Ovcha Mogila are analyzed. Both sites have a well-functioning ventilation system operates more than 4 hours a day (it turns off in the evening and turns on in the morning). To be analyzing the indoor radon daily variations in premises the spectra of direct measurements for approximately 24 hours were monitored. In the Narechenski Bani branch, radon levels have been found above the national reference level for both period of measurement. The arithmetic mean of CRn when ventilation system is functioning AM=572 Bq.m³ and when is nonoperating AM=984 Bq.m³. At these high levels it is necessary to carry out additional activities to improve ventilation (air exchange rate measurement, radon mitigation systems, and others). The comparison of radon values during running and stopped ventilation shows a statistically significant difference (MW, p<0.0001) (Fig.3).

In Ovcha Mogila are registered low levels of radon. The spa is situated away from the thermal water source and during the transport of water from the spring to the rooms degassing of radon occurs and this is the possible reason for low radon concentration [14, 15]. When the ventilation system working the AM is 18 Bq.m³ and when it is not functioning AM is 60 Bq.m⁻³. A statistically significant difference was found (MW, p=0.002) (Fig.4). The concentration of radon decreases depending on the functioning of the ventilation system, the size of the room, the meteorological conditions and, etc.



Fig.3. Distribution of the indoor radon depending on the efficient of the ventilation system in Narechenski Bani.

It is interesting to note that radon concentration decreases 3.3 times in Ovcha Mogila while radon levels in Narechenski Bani decrease 1.7 times only.



Fig.4. Distribution of the indoor radon depending on the efficient of the ventilation system in Ovcha Mogila

Increasing ventilation rate increases air change rate which decreases radon level. Direct supply of radon rich thermal water to the treatment area and non-efficient ventilation system have an impact on the radon accumulation in the air of premises. Our results find ventilation play the major role in the cumulating of radon in indoor air of spas.

DOSE ASSESSMENT

To estimate the dose in the lungs that visitors can receive in rooms we used a model of lung deposition of radon progeny and dosimetry. The effective dose of a visitor who performed procedures with thermal water in different spas was evaluated according to ICRP 137 [16]. A reference value dose coefficient of 3 mSv per mJ.h.m³ corresponds to 6.7x10⁶ mSv per Bq.h.m³ assuming an equilibrium factor, F, of 0.4 were used [16]. Normally each visitor uses the mineral water in the pools and treatment rooms from one to four hours a day. The average radon concentration was analysed from the spectra of direct measurements during the visit. The results of the analysis are presented in Table 2. The values obtained vary from 10.3 to 0.1 uSv per hour. In most of the surveyed branches (9 numbers) the values remain low and are comparable to the values reported for some spas [17, 18, 22].

 Table 2. Effective dose for visitors per hour in rooms using water

Spa	AM CRn	EfD	Humidity
	Bq.m ³	μSv	%
Momin Prohod	3852	10.3	152
Narechenski Bani	965	5.0	56
Hisarya	118	0.6	59
Pavel Banja	58	0.3	53
Varshets	61	0.3	45
Bankya	89	0.5	29
Kystendil	126	0.7	76
Sandanski	30	0.2	54
Velingrad	59	0.3	50
Ovcha Mogila	22	0.1	67
Banite	32	0.2	59

The potentially increase of air radon concentration is higher in places with water therapy, such as swimming pools, bathtubs, tangents etc. where there is a high level of steam, and humidity due to the use of more water [19]. The highest humidity is recorded in Momin Prohod, where the radon concentration is the highest and there is no ventilation system (Table 2). The technique of bath filling influenced the contribution of radon in the pool and bath areas of the spa. Other studies have found largest increase in internal radon when the bath is filled with water taps [19, 20, 21]. In our case the measurements indicate higher values during the use of geothermal water for treatment. A more accurate the radon exposure impact assessment of the environment could be done by conducting simultaneously targeted studies in both environments wet (such as spa) and dusty (such as mining).

CONCLUSION

Measurements of ²²²Rn concentrations in indoor air of spas buildings in Bulgaria were carried out using continuous measuring instruments. The radon concentration varied in the range of 16 - 4455 Bq.m³.

The visitors' effective lung dose is low (except Momin Prohod and Narechenski Bani) and is comparable with the values reported for other spa studies. The radiation dose varied depending on factors such as type of premises (with geothermal water and without), presence of a ventilation system, ventilation rate and time spent in the treatment premises.

The difference in the concentration of radon in premises with water therapy and others without water has been confirmed, which proves that geothermal water is an additional source of radon in the spa. We have also observed that mineral water as a source of radon increases the concentration of gas in the rooms where water therapy is performed.

The limited number of spas (Momin Prohod and Narechenski Bani) have radon levels in air above the national reference level. For the Narechenski Bani branch, measures must be taken to improve ventilation, while in Momin Prohod a mechanical ventilation system must be built to better protect human health.

The results of the direct radon measurements and spectrum analysis showed that working ventilation system have significant impacts on radon content in a larger number of treatment premises. One of the possible solutions for improving indoor air quality is the wellfunctioning ventilation system. The effective dose for visitors in different branches varied depending on factors such as the effective ventilation system, and the using of thermal water for therapy. The results of the study could be useful to optimize radon concentration in building with public access as spa centers.

ETHICAL APPROVAL

The authors declare that this manuscript is original, has not been published before and is not currently being considered for publication elsewhere. The results are measured and analysed by authors.

DECLARATION OF INTERESTS

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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ABREVIATIONS

222Rn - radon;

- 226Ra radium;
- CRn radon concentration, Bq.m-³;
- GM geometric mean;
- AM arithmetic mean;
- Min minimum;
- Max maximum;
- CV coefficient variation;
- EfD effective dose;
- MW Mann-Whitney test;
- KS Kolmogorov-Smirnov test.

REFERENCES

- V. Hristov, N. Stoyanov, S. Valtchev, S. Kolev, A. Benderev. IOP Conf. Series: Earth and Environmental Science, 249, 012035 (2019).
- 2. V. Hristov, M. Trayanova, S. Kolev. *National Proc. of Conference of Bulgarian Geological Society with international participation* "GEOSCIENCES 2016" (2016).
- 3. A. Benderev, V. Hristov, K. Bojadgieva, B. Mihaylova, In: Papić, P. (Ed.). Mineral and Thermal Waters of Southeastern Europe. *Switzerland, Springer International Publishing*, 47, (2016).
- 4. M. Pugliese, M. Quarto, V. Roca, *Indoor and Built Environment*, 23, 1 (2013).

- 5. UNSCEAR. Report 2006, Volume II, Annex E, *UN ed.*, *NY*, (ww.unscear.org/unscear/en/publications.html), (2009).
- 6. WHO. H. Zeeb, F. Shannoun, Eds. World *Health Organization*, (2009).
- 7. G. Manic, S. Petrovic, M. Vesna, D. Popovic, D. Todorovic, *Environm. Inter.* 32, 533 (2006).
- J. Somlai, A. Torma, P. Dombovári, N. Kávási, K. Nagy, T. Kovács, *J Radioanal Nucl Chem*, 272, 101 (2007).
- D. Nikolopoulos, E. Vogiannis, E. Petraki, A. Zisos, A. Louizi, *Sci. Tot. Environ*, 408, 495 (2010).
- 10. A.S. Silva, M.L. Dinis, Proc. of RAD Conference 2017, 5th int. conf. on radiation and application in various field of research, Budva, 2, 141, (2017).
- 11. European Commission, Council Directive 2013/59/EURATOM laying down basic safety standards for protection against the dangers arising from exposure to ionizing radiation, and repealing, *OJ of EU*, *L* 13/1. (2013),
- 12. Regulation on Radiation Protection, adopted by CM Decree № 20/14.02.2018, amended by CM No.455/22.12.2020.
- 13. E. Vogiannis, D. Nikolopoulos, *Sci. Total Environ*, **405**, 36 (2008).
- M. Inagaki, T. Koga, H. Morishima, Sh. Kimura, M. Ohta, J. Journal of Nuclear Science and Technology, 49, 531 (2012).
- M. Adelikhah, A. Shahrokhi, S. Chalupnik, E. Toth-Bodrogi, T. Kovcacs, *Iran. Heliyon*, 6 e04297 (2020).
- 16. ICRP. Occupational Intakes of Radionuclides: Part 3, Ann. ICRP 2017, 46, (2017).
- 17. V. Radolic, B. Vukovic, G. Smit, D. Stanic, J. *Environ. Radioact*, **83**, 191 (2005).
- 18. D. Nikolopoulos, E. Vogiannis, *Sci. Tot. Environ*, **373**, 82 (2007).
- M. Műllerová, J. Mazur, P. Blahušiak, D. Grządziel, K. Holý, T. Kovács, K. Kozak, E. Nagy, M. Neznal, M. Neznal, A. Shahrokhi, *Nukleonika*, 61, 303 (2016).
- E. Vogiannis, M. Niaounakis, C. P. Halvadakis, *Environ Int*, **30**, 621 (2004).
- H. Lettner, A. K. Hubmer, R. Rolle F. Steinhäusler, *Environment International*, 22, 399 (1996).
- K. Walczak, J. Olszewski, M. Zmyslony, J. of Occup. Medicine and Environmental Health, 29, 161 (2016).