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RESEARCH ARTICLE

Analysis of exposure to radon in Bulgarian rehabilitation hospitals

Kremena Ivanova[1] · **Desislava Dzhunakova**[1] · **Zdenka Stojanovska**[2] · **Jana Djounova**[1] · **Bistra Kunovska**[1] · **Nina Chobanova**[1]

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Abstract

Mineral springs are used in spa resorts throughout the world. Radon is a natural radioactive source, which can dissolve, accumulate, and be transported by water. This study investigates the radon concentration in air and water in 12 Bulgarian rehabilitation hospitals and presents the assessment of the exposure to radon in them. The measurements were performed at 401 premises within 21 buildings, using two types of passive detectors for a dry and wet environment that were exposed from February, 2019 to June, 2019. The radon concentration varied from 19 to 2550 Bq/m^3 with an arithmetic mean and a standard deviation of 102 Bq/m^3 and 191 Bq/m^3, respectively. The hypothesis that in hospitals the source of radon, besides soil under the buildings, is also the mineral water that is used for treatment was tested. Thermal water samples were procured sequentially from a spring and baths to analyse the reduction of radon concentration in them till reaching the premises. The results show that the concentration of radon decreased by approximately 50%. Further, the correlation analysis applied to the data proved the relation of the levels of indoor radon in the treatment rooms with those in the water. Mineral water used in rehabilitation hospitals have radon transfer coefficients ranging from $4.5 \cdot 10^{-4}$ to $8.4 \cdot 10^{-3}$. In addition, an analysis of the exposure of patients and workers to radon in rehabilitation hospitals based on the indoor radon levels and period of exposure was performed. The doses of workers do not exceed the limit of the annual effective dose for the population from all sources (1 mSv/year).

Keywords Mineral water · Radon · Rehabilitation hospital · Track detector · Radiation dose

Radon (^{222}Rn) is a natural radioactive gas formed from the radioactive decay of ^{226}Ra to short-lived radioactive products. Radon and its decay products are recognized as the most significant natural source of human exposure (UNSCEAR 2000), and its inhalation can cause lung cancer (WHO 2009). To identify the radon sources and explain the factors that affect radon dynamics in an indoor environment, numerous measurements of radon have been performed in various homes and workplaces around the world.

It is well known that in most of the cases, the main source of indoor radon is the radon that is generated in the underlying rock and soil of the buildings, which is transported indoors because of concentrations and pressure difference flows. In addition, the radon gas can dissolve and accumulate in water from underground sources, such as wells or mineral springs, where the water, which comes from deep springs, can contain high radon concentration because of leaching of rocks, making it an additional source of indoor radon. Further, rehabilitation centres use mineral water for therapy, which can have higher levels of radon. Furthermore, concentrations of radon (^{222}Rn) in thermal waters can vary from 10 to above 1000 Bq/l (Szerbin 1996; Vogiannis et al. 2004; Manic et al. 2006; Nikolopoulos et al. 2010). The balneotherapy process using thermal water contributes to radon release into the indoor air and because of large volume of water used, the concentrations could reach a high value. Considering the health effects of radon, the professional staff could be exposed to a significant amount of radon. In literature, the annual effective doses reported for such workers have varied from several units to tens mSv per year (Radolić et al. 2005; Žunić et al. 2006). The problem of the radon in

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✉ Kremena Ivanova
k.ivanova@ncrrp.org; kivanova1968@gmail.com

[1] National Centre of Radiobiology and Radiation Protection, 3 Sv. Georgi Sofiyski St., 1606 Sofia, Bulgaria

[2] Faculty of Medical Sciences, Goce Delcev University of Stip, 10-A Krste Misirkov st., 2000 Stip, Republic of North Macedonia

air and water in thermal spas have been discussed by lot of author in the last years as the results of recent studies (Çetinkaya and Biçer 2021; Santamarta et al. 2020; Erdogan et al. 2020; Silva et al. 2020; Gulan et al. 2020; Turtiainen et al. 2021; Deeba et al. 2020; Chen and Harley 2020).

The Council of the European Union issued the European Basic Safety Standards (EU BSS) in December 2013, which recommended that the annual average radon concentration should not be higher than 300 Bq/m^3 in dwellings, workplaces and building with public access (EC 2013).

Bulgaria has an abundance of mineral water springs located throughout its territory (Hristov et al. 2016). According to Vassileva (1996), the number of mineral springs in Bulgaria is over 520, with different compositions, temperatures and properties of mineral water with most of them being warm. Fifty-seven of the 230 existing spas are promoted as balneological resorts (Hristov et al. 2019). There have been certain investigations of natural radioactivity and radon in mineral water in the literature, which shows the presence of a high concentration (Kamenova-Totzeva et al. 2018). This implies that in spa or in rehabilitation centres, staff and patients may have been exposed to higher levels of radon, leading to significant additional exposure. Although Bulgaria has adapted its radiation protection standards based on the European directive, there exists no systematic indoor radon measurements in public buildings where mineral water is used.

This paper focused on the results of indoor radon (C_{Rn}) and radon in water (C_{Rnw}) measurements obtained via a survey of 12 thermal specialized hospitals for rehabilitation in Bulgaria. The aim of the survey, realized in the framework of a project funded by Bulgarian National Science Fund, was to investigate radon level in air and water and relation between them. The hypothesis of availability of an additional radon source for treatment premises and assessment of exposure was explored.

Materials and method

Objects and design of survey

Bulgaria is situated in the north-eastern part of the Balkan Peninsula. Several thermal and mineral water springs exist within the Bulgarian territory with great variation in the physical properties and chemical composition because of the diverse geological structure of the country. The use of thermal water for treatment in Bulgaria has been known since ancient times. Specialized rehabilitation hospitals were established in the 1960s, and in the past, there were more hospitals. Currently, there are 13 branches left, where the treatment takes place. The surveyed branches are described in Table 1, where the codes, administrative location (settlement, municipality and district) and number of measured premises are presented. Measurements were not performed at the Pomorie branch located on the Black Sea coast, as it uses healing sea mud instead of mineral water.

All the branches (11) of the national complex are located in mountainous regions, except for one present in the Danube plain (the village of Ovcha Mogila). The locations within the territory of Bulgaria are presented in Fig. 1. The specialized hospitals are located primarily in small spa resorts, with only Kyustendil being a relatively large town and the administrative centre of the district. Five of these specialized hospitals are situated within one building, while the rest are in two or three buildings. The bathrooms with pools in the branch at

Table 1 Location of surveyed specialized rehabilitation hospitals with their code, number of buildings, measured premises with and without using water for the treatment and percentage of losses of detectors

Code	Location (village, municipality, district)	No. of buildings	No. of premises	No. of premises for water treatments	% of detectors losses
H1	Narechenski Bani, Asenovgrad, Plovdiv	1	19	10	9
H2	Momin Prohod, Kostenec, Sofia-district	2	27	22	14
H3	Banya, Karlovo, Plovdiv	3	12	9	40
H4	Hissarya, Hissarya, Plovdiv	1	23	7	14
H5	Pavel Banya, Stara Zagora	2	31	15	12
H6	Varshets, Montana	2	33	15	11
H7	Bankya, Sofia, Sofia-city	1	18	2	0
H8	Kyustendil, Kyustendil	2	21	5	19
H9	Sandanski, Blagoevgrad	1	18	5	0
H10	Velingrad, Pazardjik	3	43	8	42
H11	Ovcha mogila, Svishtov, Veliko Tarnovo	1	24	6	0
H12	Banite, Smolian	2	23	6	6
Total		**21**	**291**	**110**	**18**

Fig. 1 Location of the surveyed specialized rehabilitation hospitals with their codes

Banya-Karlovo are situated in two separate small buildings on one floor. Further, an additional building of the Ministry of Health was investigated in Velingrad, which was also used for rehabilitation purposes. The exact number of buildings is presented in Table 1. The buildings are usually large having several floors (from 2 in Pavel Banya and Varshets to 9 in Narechen) and the total area of these buildings varies from 360 to 8500 m^2. The underground floors in the hospitals are usually occupied with, a swimming pool or treatment rooms situated in them, but in some of hospitals, the underground floor is used for storage of mineral water in tanks or for warehouses and workshops.

Questionnaires to gather information about the type of building construction, year of construction and repair, way of use of premises, way of heating and other information were handed out and collected. In some mineral deposits, such as Hissarya, Banya-Karlovo, Varshets, Velingrad and Narechen, there are several thermal springs with different compositions and properties. Typically, the water from one such spring is used in the hospitals located at these places for treatment: in Banya and Karlovo water is utilized from three springs, while in Velingrad two springs are used.

The indoor radon (C_{Rn}) in treatment rooms, pools and offices on the basement and ground floor were measured with passive detectors. Measurements were also performed on the first, second and upper floors, but not in all premises. Radon CR-39 detectors in premises without water procedure were positioned on a shelf at a distance of approximately 1–1.5 m above the floor or any wall surface. In premises where water is used, such as swimming pools and bathrooms, the detectors were suspended via a rope from the ceiling or mounted on partitions separating showers, bathtubs or treatment rooms. For the measurements of radon concentration in water (C_{Rnw}), samples were procured from the spring (borehole or casing) and from the treatment rooms (baths). The number of detectors, being divided into rooms in which water is used for treatment and ordinary premises (offices, doctors' offices, rooms), is presented in Table 1. In the rehabilitation hospitals, as with public facilities, there are losses of detectors because they are visited by many people, and at the end of the exposure period, several of the detectors were missing. Higher losses were expected because the surveyed buildings are used by a lot of peoples. The total losses amounted to 18%, and the distribution by branches is summarised in Table 1. The biggest losses were faced at the branches of Banya, Karlovo and Velingrad. The bathrooms, in these branches, are present in separate buildings and are visited by more people, which could explain the losses. Thus, the total number of analysing detectors was 401, with 291 detectors on premises where water was not used for treatment and 110 in premises with water procedures present, such as pools, baths and procedure rooms.

Indoor radon measurement

The indoor radon concentration (C_{Rn}) measurements in the premises of the hospital were performed using RSKS-type and RSFW-water protected type nuclear track detectors produced by Radosys. The RSKS-type detectors consisted of one CR-39 radon-sensitive chip, while RSFW had two chips, enclosed in diffusion chambers. The latter has protection from high humidity and is specifically developed for conducting measurements of C_{Rn} in the premises of spas, caves and mines. The detectors have a unique ID from the manufacturer, which was used to identify its movement from the laboratory to the room where the measurement was being performed and vice versa. Sampling, processing and calculation of results were performed in accordance with ISO 11665–4 (2012). Further, the detectors from each batch were exposed to a standard radon atmosphere in an accredited laboratory in BfS Germany, and the determination of the calibration coefficient (F_c) for each batch was performed in the laboratory of the National Center for Radiobiology and Radiation Protection in a manner similar to the processing of the detectors. The relative uncertainty of the calibration combined with the declared uncertainty of the accredited laboratory resulted in an estimated relative stationary deviation of the transit and exposure detectors of 3%. Further, the uncertainty was determinate considering the probability distribution of the results. The radon concentration was obtained by following the steps described in the ISO standard as well as applying equestrians for determination of activity and standard uncertainty.

Using Eq. 1 the indoor radon concentration C_{Rn} is determined due to calibration factor (F_c), net track density ($d-b$) and period of detector exposure (t):

$$C_{Rn} = F_C \bullet (d - b) \bullet t^{-1} \tag{1}$$

where d and b are the counted and background track density (number of tracks per unit area), respectively.

The relative combined uncertainty of result is determined by the formula as follows:

$$u\left(C_{Rn}\right) = C_{\mathrm{Rn}} \bullet \sqrt{\left(\frac{u(F_{\mathrm{c}})}{F_{\mathrm{c}}}\right)^2 + \left(\frac{u(d)^2 + u(b)^2}{(\bar{d} - \bar{b})^2}\right) + \left(\frac{u(t)}{t}\right)^2} \tag{2}$$

where $u(F\mathrm{c})$, $u(d)$ and $u(b)$ are the uncertainties of the calibration factor, standard deviation of the track density reading (d) (estimated by the microscope) and standard deviation of track density of the background group (b) of 10 detectors, respectively. Further, the uncertainty of the exposure period assuming a triangular distribution (Stojanovska et al. 2017) was determined as $u(t) = 1/\sqrt{6}$. Thus, the combined standard uncertainty of the method was assessed at 10% (at 95% confidence level). The minimum detectable concentration was 12 Bq/m^3 for RSFW-water protected type and 18 Bq/m^3 for RSKS-type. The detectors were deployed in the premises of the specialized hospitals in February, 2019 and collected in June 2019. In addition, the conservative assumption that the results of the indoor radon concentration obtained through the detector exposure over a period of 6 months (covering the winter and the spring season) represented the annual radon concentrations was considered in this study (Stojanovska et al. 2016).

Sampling and measurement of radon in water

The method based on Passive Environmental Radon Monitor (E-PERM) Electret Ion Chamber (EIC) system manufactured by Rad Elec Inc. was used for the measurement of the radon concentration in water, which consists of a reader, S-chambers and long-term (LT) or short-term (ST) electret, jars of 3.72 l volume and sample bottles of 68 and 136 ml. The water samples were collected using 136-ml collection bottles. Further, a bucket was used to till it overflowed, after the water ran for several minutes and, thereafter, raised such that the tap (or source) was below the surface. The sampling bottle was subsequently submerged and filled from the bottom of the bucket and capped underwater (Kotrappa 1999). Three parallel bottles from the sampling point were procured. Two of them were measured simultaneously, while the third was a control sample and measured when the results of the first two differed. The notes for exact data and hour, ID of sample and duration of transport to laboratory were maintained. In the laboratory, the sampling bottle was placed without cap at the bottom of the glass jar, and an E-PERM short-term chamber with a long-term electret or configuration of ionizing chamber (SLT) was suspended over the water spread on the bottom. Subsequently, the jar was sealed and remained closed for 24–48 h, and the exact duration of period was written in protocol of the sample. The evaluation of radon concentration in water (C_{RnW}) was based on radon concentration in air inside jar C_{Rnj}, normalized to the volume of water sample and the jar and two periods (T_{d}, the period utilized for collection of samples till immediately before insertion of the sample for analysis, and T_{a}, the period utilized for inserting the sampling bottle into the measuring jar till the ionizing chamber which is removed for analysis). The measurement of C_{Rnj} inside the jar was estimated under the voltage difference before and after exposure of detector in jar, gamma dose rate in laboratory and appropriate calibration factors, based on the procedure described by the producer (Rad. Elec. Inc., Frederick, MD, USA). Therefore, the C_{RnW} was calculated after measuring the C_{Rnj} by multiplying by the constants C_1, C_2 and C_3 as follows:

$$C_{\mathrm{Rnw}} = C_{\mathrm{Rnj}} \bullet C_1 \bullet C_2 \bullet C_3 \tag{3}$$

where the constant C_1 considers the delay period between the collection of the water sample and the beginning of the measurement and is expressed as.

$$C_1 = e^{(\lambda \bullet T_{\mathrm{d}})}, \text{ as } \lambda = 0.1814 \tag{4}$$

The constant C_2 is the constant based on the analysis period and is expressed as

$$C_2 = C_2 = \frac{\lambda \bullet T_{\mathrm{a}}}{1 - e^{(-\lambda \bullet T_{\mathrm{a}})}} \tag{5}$$

The constant C_3 is the ratio between the volume of the jar and that of the sampling bottle, and this case was equal to 28. In addition, the reader was regularly sent to the producer for calibration. The method was validated with the encapsulated ^{226}Ra source in water during the intercomparison (Kitto et al. 2010), and the estimated combined uncertainties of the method was 20% (at 95% confidence level).

Data analysis

The results obtained were evaluated and systematized, and the information from the completed questionnaires was summarised for analysis. This was achieved by employing the SPSS (version 19) statistical software. In the analysis, depending on the distribution and homogeneity of the grouped C_{Rn} results, appropriate parametric and non-parametric statistical tests were used at a 95% confidence level. Based on the data, the average transfer factor was set as the ratio of the radon concentration in the air to that in water in the treatment premises ($f_{\mathrm{t}} = C_{\mathrm{Rn}}/C_{\mathrm{Rnw}}$), which describes the transfer of radon from water to air. Although the transfer factor depends on the water temperature (Cosma et al. 2008), in this study, the influence of temperature was neglected because the water temperature in the pools and baths of the hospitals were maintained at approximately the same temperature (approximately 30 °C). Depending on the temperature of the water in the springs, it is heated or cooled before being introduced into the baths.

To assess the radon exposure of patients and workers in rehabilitation hospitals, the annual effective dose (E) due to inhalation of radon was estimated according the EU Radiation Protection N° 193 (2020) as follows:

$$E = C_{\mathrm{Rn}} \bullet F \bullet t \bullet C_f \tag{6}$$

where E is expressed in mSv, C_{Rn} is the average indoor radon concentration in Bq/m^3 in premises with water procedure and F is the equilibrium factor between radon gas and its decay product. The value of the equilibrium factor, F, depends primarily on the indoor ventilation rate because of opening/shutting of windows and the use of electric fans and air conditioners. Although the equilibrium factor in thermal spas, according to some authors, varies from 0.2 (Vogiannis et al. 2004) to 0.6 (Soto and Gómez 1999), the standard assumption of $F = 0.4$ was applied for calculation (UNSCEAR 2000; ICRP 2017). Further, t is exposure time in hours. Effective doses of inhalation of radon in rehabilitation hospitals by a reference worker and patient were calculated assuming the breathing rate of 1.2 m^3 h^{-1}, that is, approximately one-third of time spent sitting and two-thirds of time spent in light exercise. In addition, for the calculation of radon effective doses, a dose coefficient of 3 mSv per mJ h m^{-3} (approximately 10 mSv/WLM)corresponding to $C_{\mathrm{f}} = 6.7\ 10^{-6}$ mSv/[(Bq.h/m^3)] was applied (ICRP 2017). The dose coefficients were calculated using defined biokinetic and dosimetric models for reference person under particular exposure conditions as reference values and were not regarded as subject to uncertainty (ICRP 2007). The sources of uncertainties in biokinetic models are associated with the types of information used to construct the models for the elements. Furthermore, the uncertainty in the dose assessment depends on uncertainties associated with measurements of radon concentration and sampling and uncertainties in the exposure scenario, including factors such as period of exposure. However, for regulatory purposes, the models and parameter values were fixed by convention.

Results and discussion

Radon concentration in air

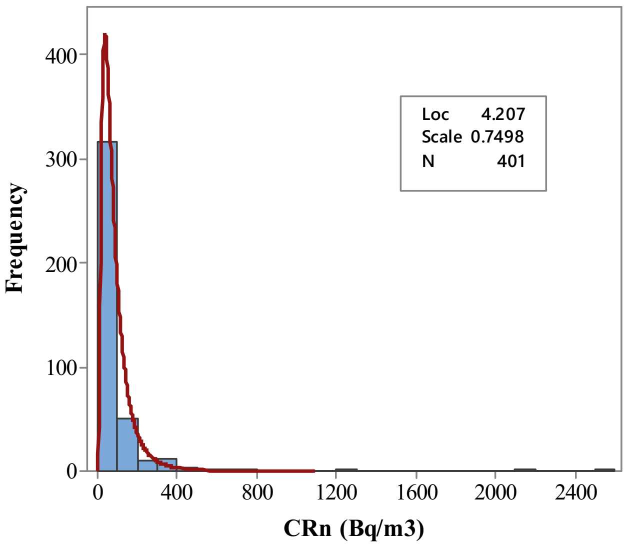

Fig. 2 Histogram of C_{Rn} fitted with log normal function

Figure 2 shows the histogram of C_{Rn}, measured in all premises considered in this study. It is noted that the C_{Rn} are generally lower, with the exception of 27 rooms in which the C_{Rn} are higher than reference level of 300 Bq/m^3 as set by national legislation. In order to normalize the C_{Rn} data, the values were ln transformed. The hypothesis that $\ln C_{\mathrm{Rn}}$ have a normal distribution was not confirmed at 95% confidence level.

To evaluate the spa region, the measured buildings were grouped by location, and Table 2 presents the descriptive statistic and p value of Shapiro–Wilk test for normality of $\ln C_{\mathrm{Rn}}$ distribution ($p > 0.05$, 95% confidence level). Considering the p values in Table 2, it follows that the ln-transformed results from most spa regions follow a normal distribution or the data are in the vicinity the mean value more often. However, the data of Narechenski Bani (H1), Pavel Banya (H5), Varshets (H6) and Velingrad (H10) spa regions do not exhibit a normal distribution. However, to avoid the influence of extreme values, all further analyses were performed on $\ln C_{\mathrm{Rn}}$. The reasons for inhomogeneity of the C_{Rn} can be many. The results of C_{Rn} are influenced by differences in geology between locations but also differences in premises. As a result, there is a wide range of C_{Rn} values between the locations and on the locations themselves. The measured C_{Rn} have a minimum value of 19 Bq/m^3 in the Velingrad branch (H10) up to 2550 Bq/m^3 in. The significant difference between C_{Rn} in the different spa regions was confirmed by the Kruskal–Wallis test (KW, $p < 0.0001$). Furthermore, we tested the difference between C_{Rn} in each two regions separately via the Mann–Whitney test, which conforms to the assumption of their grouping Fig. 3. We find all differences are significant with $p < 0.05$ between the locations:

The highest C_{Rn} was found at Momin Prohod (H2), which implies that it is a spa region with high radon. The variation coefficient was high too, indicating the high range of C_{Rn}. The average C_{Rn} of measured buildings in Momin Prohod were found higher than reference level of 300 Bq/m^3. In a

Table 2 Descriptive statistics radon concentrations by location: *N* number of measurements, *AM* arithmetic mean, *SDV* standard deviation, *Min.* minimum, *Max.* maximum, *CV* coefficient of variation, *GM* geometric mean and *p* value of the Shapiro–Wilk test for normality

Code of location	Indoor radon concentration (C_{Rn})							
	N	*AM*, Bq/m^3	*SDV*, Bq/m^3	Min, Bq/m^3	Max, Bq/m^3	*CV*, %	*GM*, Bq/m^3	Shapiro–Wilk, *p**
H1	29	128.1	140.9	35	753	110	98	0.003
H2	48	335.7	482.3	37	2550	144	196	0.061
H3	21	103.9	58.1	45	305	56	93	0.358
H4	30	67.5	38.4	21	176	57	58	0.568
H5	46	111.6	76.3	33	392	68	95	0.052
H6	48	67.5	32.9	32	237	49	62	0.007
H7	20	50.4	21.1	22	91	42	46	0.424
H8	26	37.0	11.0	23	60	30	35	0.145
H9	23	30.9	7.3	21	47	24	30	0.447
H10	51	51.7	28.1	19	117	54	45	0.011
H11	30	60.3	22.6	33	120	37	57	0.284
H12	29	51.4	19.3	24	90	37	48	0.267
Total	401	102.3	195.3	19	2550	191	67	<0.0001

*$p > 0.05$ (95% confidence level)

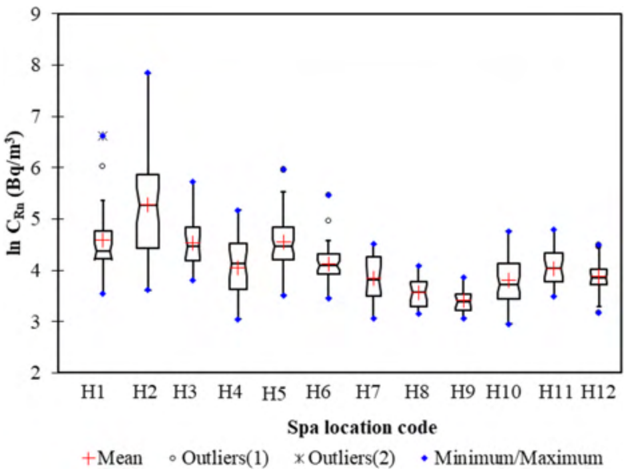

Fig. 3 The boxplots of C_{Rn} distribution by location of the surveyed specialized rehabilitation hospitals

similar manner, C_{Rn} values were found to be high for the region of Niska Banja, Serbia by Žunić et al. (2006) and in Portuguese thermal spas (Silva et al. 2020). In addition, the results of a survey conducted in Spain show a C_{Rn} of over 5000 Bq/m^3 (Soto et al. 1995), which were higher than the results in Momin Prohod. Further, Narechen (H1), Banya-Karlovo (H3) and Pavel Banya (H5) were found to be the spa regions with moderate C_{Rn} values. These regions need to be further investigated in detail, and actions are required to inform the public in accordance with the national radon plan. The arithmetic mean value of C_{Rn} in the region with low radon in the range from 30.9 Bq/m^3 in Sandanski (H9) to 37 Bq/m^3 in Kyustendil (H8). The coefficient of variation, which depicts the extent of variability in relation to the mean of the data in those regions, was also relatively small. The results from low radon spa region are in the similar variation range, as in the thermal baths of Rimini and Pesaro-Urbino provinces, Central Eastern Italy (7–71 Bq/m^3), published by Desideri et al (2004). Moreover, the C_{Rn} in five Slovenian spas, at Rogaska Slatina, Radenci, Moravci, Podcetrtek and Catez, were in the same low range, because of the effective ventilation systems (Vaupoti and Kobal 2001).

For a more detailed assessment of the radon variations in the buildings, the results were grouped based on the type of premises, whether water was used for treatment and room types such as ordinary rooms or offices on the underground and ground floor only.

The arithmetic mean value for the procedure rooms was calculated to be $AM = 186$ Bq/m^3, which was approximately three times higher than in rooms without water procedures ($AM = 77$ Bq/m^3). Subsequently, a non-parametric Mann–Whitney test was applied for the two groups of rooms and a statistically significant difference (MW, $p < 0.001$) was found. Figure 4 shows the comparison of the results between the premises with water procedures (treatment rooms) to those without them.

For further assessment of the distribution of radon concentration by floors, the results are grouped into four groups as follows: underground, ground floor, first floor and above the first floor (Fig. 5).

Subsequently, a non-parametric Kruskal–Wallis test was applied and a statistically significant difference between the groups (KW, $p < 0.001$) was confirmed. To further check between which groups a difference existed, the Mann–Whitney rank test was used for all couples, and no statistically

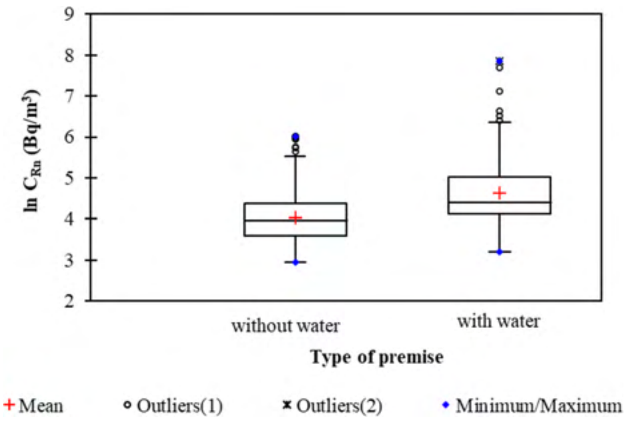

Fig. 4 The boxplots of C_{Rn} by type of premises in the surveyed specialized rehabilitation hospitals according the used of water in them

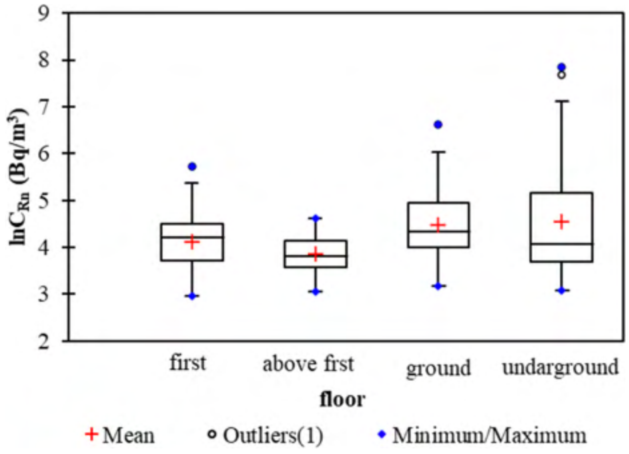

Fig. 5 The boxplots of $\ln C_{Rn}$ by floors in the surveyed specialized rehabilitation hospitals

significant difference was observed between the underground, ground and first floor. However, a difference was found between the group with results above the first floor and the lower floors (MW, $p < 0.001$) which can be explained by the location of most treatment premises on the lower floors. This finding suggests that water could be a significant source of radon in most premises. The same conclusion was made by Santamarta et al. (2020). It can be noticed from Fig. 6, the C_{Rn} in the underground rooms without water treatment are much lower than in the treatment rooms where the water further increases the C_{Rn}. Given that the most treatment rooms are located on the ground floor, we assume that radon from the water further increases the indoor concentration in all rooms. On the first and upper floors, the effect of geogenic radon weakens, and the C_{Rn} decreases in rooms without treatments. In the treatment rooms on those floors, the C_{Rn} is almost the same, indicating that water is an additional source of indoor radon.

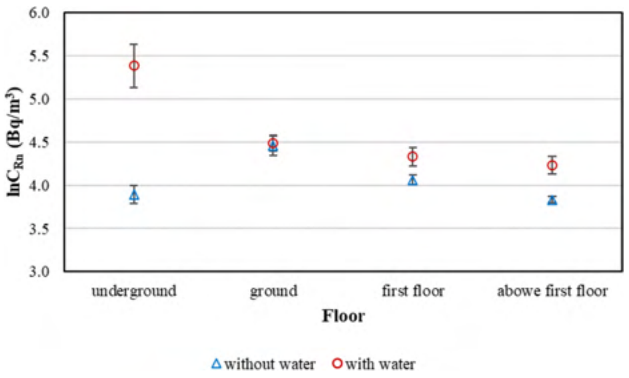

Fig. 6 The mean values of $\ln C_{Rn}$ grouped by the type of premise and floor

Radon concentration in water

The results of the concentration of radon in the mineral water from springs and baths by location are presented in Table 3. The measurements show that the concentration of radon in the mineral water taken from the baths in the hospitals has decreased up to 50% till the water reaches the bath premises. This was expected, as in some places the water collects in tanks or reaches the hospital through pipes, where the radon emanates or decays to its daughter products. For each branch, the arithmetic mean in the treatment rooms was estimated and is presented in Table 3.

In the rehabilitation hospitals at Banya, Karlovo (H3) and Velingrad (H10), thermal water is used directly in the bathroom from the spring. The C_{RnW} from the springs at Narechenski Bani (H1), Momin Prohod (H2), Pavel Banya (H5) and Velingrad (H10) were above the standards set by both the European Union reference level set at 100 Bq/l (EU 2001) and limit for drinking water as imposed by the Bulgarian regulation. Such high values of C_{RnW} were reported by many other authors of several countries, as in Venezuela (Pugliese et al. 2014), Niška Banja Serbia (Žunić et al. 2006) and Spain (Soto et al. 1995). Further, the calculated radon transfer coefficients are presented in Table 3. The range of the transfer coefficients were similar to those in Stubica, Croatia (Radolić et al. 2005), but in most hospital bathrooms, it was higher than the typical estimated values from shower for normal water of approximately 10^{-4} (Nazaroff and Nero 1988; Vinson et al. 2008).

Spearman's rank correlation coefficient was used to assess the relationship between the concentration of radon in mineral water and in indoor air. The test is statistically significant ($p < 0.05$ at 95% confidence interval) and Spearman's correlation coefficient between the C_{RnW} from baths and C_{Rn} was $\rho_o = 0.806$. The correlation analysis showed that 65% ($\rho^2 = 0.65$) of radon measurements in air can be explained by the presence of radon in bath water, that is, high values

Table 3 Radon concentrations in springs and bath water, average radon concentration in bath air and treatment premises and their respective transfer coefficients

Specialized hospital for rehabilitation (code of location)	Temperature of water, °C	C_{RnW}, Bq/l (spring)	C_{RnW}, Bq/l (bath)	*AM* C_{Rn}, Bq/m^3 (treatment premises)	Transfer coefficient
Narechenski Bani(H1)	28	613	189	186	$9.8\cdot10^{-3}$
Momin Prohod (H2)	64.5	1265	270	795	$2.9\cdot10^{-3}$
Banya, Karlovo (H3) — men's bath	25	19		125	$6.6\cdot10^{-3}$
Banya, Karlovo (H3) — women's bath	26	17		138	$8.1\cdot10^{-3}$
Banya, Karlovo (H3) — central spring	34,5	58		166	$2.9\cdot10^{-3}$
Hissarya (H4)	45	30	15	62	$4.1\cdot10^{-3}$
Pavel Banya (H5)	61	428	223	95	$4.3\cdot10^{-4}$
Varshets (H6)	37,6	22	31	72	$2.32\cdot10^{-3}$
Bankya (H7)	23	24	4.3	36	$8.37\cdot10^{-3}$
Kyustendil (H8)	73	6	11	45	$4.02\cdot10^{-3}$
Sandanski (H9)	72	30	23	28	$1.21\cdot10^{-3}$
Velingrad (H10) — Kamena spring	47	222		86	$3.87\cdot10^{-4}$
Velingrad (H10) — Veliova bania spring	63	110		135	$1.23\cdot10^{-3}$
Ovcha mogila (H11)	45	159	32	77	$2.41\cdot10^{-3}$
Banite, Smolian (H12)	42	14	16	78	$4.88\cdot10^{-3}$

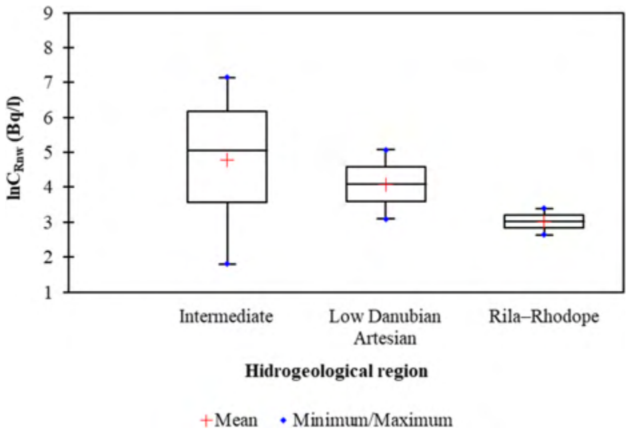

Fig. 7 The boxplots of lnC_{RnW} from springs by hydrogeological regions in Bulgaria

of radon in the air could be expected to be found if there are high values of radon in the water. To systematize the results and link them to hydrogeology, the C_{RnW} results from the springs were grouped by the hydrogeological regions of the country. Bulgaria has been divided into three major hydrogeological regions, which is closely linked to the main geological structures, namely, low Danubian Artesian, intermediate and Rila-Rhodope (Benderev et al. 2016; Hristov et al. 2019).

Results of radon concentration in water from springs by their hydrogeological region are presented on Fig. 7. The lowest arithmetic mean of the C_{RnW} from springs (25.7 Bq/l) was found for the intermediate hydrogeological region, while the highest was for the Rila-Rhodope region (375.5 Bq/l). There was no statistically significant difference in the results of the C_{RnW} by their hydrogeological regions, which indicates that radon in water may be influenced by additional factors and a more detailed study of the geology of spa areas should be performed.

Estimation of exposure to radon

Based on the average C_{Rn} in the premises and Eq. 4, the average annual effective dose to workers and patients because of radon exposure was estimated. Depending on the place of work, the doses of workers were evaluated for premises with water procedures and those without. In addition, the effective dose depends on the exposure period, and it was estimated based on the time that the persons stay on the premises.

The main uncertainty in calculation of effective dose was the exposure period, which was applied. The working hours in the premises with water therapy, such as swimming pools, bathtubs, tangents and others have been reduced and are 1750 h per year. In contrast, at the other workplaces in the rehabilitation hospitals the working hours are 2000 h a year. To calculate the additional annual effective dose (*E*, μSv/y) of a patient receiving a full treatment programme, it was considered that each patient receives 30 treatments annually (1 month) of 1-h duration in the pool, bath or other premises with water. The estimated effective doses from radon inhalation by specialized hospitals for rehabilitation of workers and patients are presented in Table 4. The effective doses because of radon inhalation of workers in premises with water were higher and varied from 0.3 to 14.4 mSv/y

Table 4 Estimated effective doses from radon inhalation by specialized hospital for rehabilitation of workers and patients

Specialized hospital for rehabilitation (Code of location)	Dose of public (patients), *E*, mSv/year	Dose of workers in premises with water, *E*, mSv/year	Dose of workers in premises without water, *E*, mSv/year
Narechenski Bani(H1)	0.15	8.8	1.3
Momin Prohod (H2)	0.25	14.4	2.0
Banya, Karlovo (H3)	0.06	2.0	1.0
Hissarya (H4)	0.02	0.7	0.9
Pavel Banya (H5)	0.04	1.1	1.6
Varshets (H6)	0.03	0.8	0.9
Bankya (H7)	0.01	0.4	0.7
Kyustendil (H8)	0.01	0.5	0.5
Sandanski (H9)	0.01	0.3	0.4
Velingrad (H10)	0.01	1.6	0.6
Ofcha mogila (H11)	0.02	0.9	0.8
Banite, Smolian (H12)	0.02	0.9	0.6

depending to radon concentration in rehabilitation hospitals. As expected, the workers in the rehabilitation hospital at Momin Prohod received the highest dose. The effective doses of the workers were in range of the assessed doses of workers in the thermal spas of Ischia Island (Pugliese et al. 2014) and lower than the maximum value (4.36 mSv) estimated for workers in Ilgin thermal baths (Erdogan et al. 2020). In contrast, the estimated effective doses of radon inhalation for patients are relatively low and range from 2.0 to 250 μSv/year. Though these doses do not exceed the limit of the annual effective dose for the population from all sources (1 mSv/year), in some regions (Narechenski Bani and Momin Prohod), they were higher than the levels of exemption of 10 μSv/year according to the Bulgarian legislation standards.

Conclusion

Radon measurements in the air and geothermal water of 12 Bulgarian rehabilitation hospitals were performed, and the indoor radon concentrations and water of the springs and baths were obtained in the range of 19 to 2550 Bq/m^3, 6 to 1265 Bq/l and 4.3 to 270 Bq/l, respectively. The correlation analysis was used to test relationships between the data of the concentration of radon in the mineral water and in the air in the treatment rooms and the Spearman's rank correlation coefficient was calculated to be $\rho = 0.806$, which proves that the connection of the levels of indoor radon is related to those of the water. Further, the difference in radon concentration in the premises with water therapy and those without was confirmed. The results clearly show that thermal water with high radon concentration is the source of radon in buildings. In addition, the analysis and assessment of exposure to radon confirmed the need of radon control in spas to adhere to the European Basic Safety Standard (EU BSS, Council Directive 2013/59/Euratom, 2014), which could be realized by inspecting the ventilation system, an important part for improving the indoor environment in spa. The five spa regions were identified according the indoor radon concentration which could be used to apply a graded approach in the control of spas. Further, an analysis of the results of radon concentration in water from springs by their hydrogeological regions was performed. Intermediate hydrogeological region had the lowest arithmetic means of the C_{RnW} from springs ($C_{RnW} = 25.7$ Bq/l), while Rila-Rhodope region had the highest ($C_{RnW} = 375.5$ Bq/l). Further detailed study of the geology of spa areas should be performed for assessment of the factors that influence the radon concentration in water. Finally, the estimation of annual effective doses for workers and patients were evaluated considering the exposure period and the measured indoor radon concentration. Although the study was done in only 12 regions with mineral water in the specialized rehabilitation hospitals, the results concerning the indoor radon and water variation could be used to optimize future radon surveys in buildings with public access as spa centres.

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Availability of data and materials The datasets generated and analysed during the current study are available in the NCRRP database and are available from the corresponding author on reasonable request. The data reports are publicly available on the www.radon.bg.

Declarations

Ethics approval and consent to participate We declare that this manuscript is original, has not been published before and is not currently being considered for publication elsewhere. No data, text or theories by others authors are presented, except the citations on manuscript. The results are measured and analysed by authors of the manuscript. Our manuscript is not involving human participants, human data or human tissue and animals.

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References

Benderev A, Hristov V, Bojadgieva K, Mihailova B (2016) Thermal waters in Bulgaria. In: Environmental Earth Sciences. Springer; Cham, pp 47–64

Çetinkaya H, Biçer M (2021) Radon and radium measurements of Eskişehir spa waters. Radiation Protection Dosimetry

Chen J, Harley NH (2020) A review of radon equilibrium factors in underground mines, caves, and thermal spas. Health Phys 119(3):342–350

Cosma C, Moldovan M, Dicu T, Kovacs T (2008) Radon in water from Transylvania (Romania). Radiat Meas 43(8):1423–1428

Deeba F, Rahman SH, Kabir MZ (2020) Radon concentration in soil and groundwater of west coastal area, Bangladesh. Radiat Prot Dosim 191(3):341–348

Desideri D, Bruno MR, Roselli C (2004) 222 Rn determination in some thermal baths of a central eastern Italian area. J Radioanal Nucl Chem 261(1):37–41

Erdogan M, Abaka M, Manisa K, Bircan H, Kus C, Zedef V (2020) Indoor radon activity concentration and effective dose rates at schools and thermal spas of Ilgin. Nucl Technol Radiat Prot 35(4):339–346

European Commission, (2001) European Union Commission Recommendation on the protection of the public against exposure to radon in drinking water supplies. Office Journal of the European Community, L 344, 28 December, 85–88

European Commission (2013) Laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, and repealing, Council Directive 2013/59/EURATOM. OJ of EU, L13/1

European Commission (2020) Radiation protection n° 193 Radon in workplaces, Directorate-General for Energy Directorate D—Nuclear Energy, Safety and ITER Unit. Radiation Protection and Nuclear Safety, D3

Gulan L, Penjišević I, Stajic JM, Milenkovic B, Zeremski T, Stevanović V, Valjarević A (2020) Spa environments in central Serbia: Geothermal potential, radioactivity, heavy metals and PAHs. Chemosphere 242:125171

Hristov V, Trayanova M, Kolev S (2016) Mineral water and geothermal energy application in Bulgaria. National Conference with international participation “GEOSCIENCES 2016”, Bulgarian Geological Society, 149–150 (in Bulgarian)

Hristov V, Stoyanov N, Valtchev S, Kolev S, Benderev A (2019) Utilization of low enthalpy geothermal energy in Bulgaria. In IOP Conference Series. Earth and Environmental Science (vol 249, no. 1, p 012035). IOP Publishing

International Commission on Radiological Protection (2007) Protection, radiological. Annals of the ICRP, 37, ICRP Publication, 2(4), 2, 103

International Commission on Radiological Protection (2017) Occupational intakes of radionuclides: Part 3. Annals of the ICRP, 46. ICRP Publication, 3/4, 137

International Organization for Standardization (2012) Measurement of radioactivity in the environment—air: radon-222. Part 4: integrated measurement methods for the determination of the average radon activity concentration in the atmospheric environment using passive sampling and delayed analysis. ISO 11665–4:2012

Kamenova-Totzeva RM, Totzev AV, Kotova RM, Ivanova-Teneva GR (2018) Quantitative and qualitative study of radon content in Bulgarian mineral waters. Radiat Prot Dosimetry 181(1):48–51

Kitto ME, Menia TA, Bari A, Fielman EM, Haines DK (2010) Development and intercomparison of a reusable radon-in-water standard. Radiat Meas 45(2):231–233

Kotrappa P (1999) E-PERM electret ion chambers for measuring radon in water. E-PERM System Manual. Rad Elec Inc., pp 1–13

Manic G, Petrovic S, Vesna M, Popovic D, Todorovic D (2006) Radon concentrations in a spa in Serbia. Environ Int 32(4):533–537

Nazaroff WW, Nero AV (1988) Radon and its decay products in indoor air

Nikolopoulos D, Vogiannis E, Petraki E, Zisos A, Louizi A (2010) Investigation of the exposure to radon and progeny in the thermal spas of Loutraki (Attica-Greece): results from measurements and modelling. Sci Total Environ 408(3):495–504

Pugliese M, Quarto M, Roca V (2014) Radon concentrations in air and water in the thermal spas of Ischia island. Indoor Built Environ 23(6):823–827

Radolić V, Vuković B, Šmit G, Stanić D, Planinić J (2005) Radon in the spas of Croatia. J Environ Radioact 83(2):191–198

Santamarta JC, Hernández-Gutiérrez LE, Rodríguez-Martín J, Díaz RM, Bascones RJL, Moro ÁMG, Cruz-Pérez N (2020) Radon measurements in water galleries in Tenerife, Canary Islands (Spain). Air Qual Atmos Health 13(11):1287–1292

Silva AS, Dinis MDL, Pereira A (2020) Indoor radon levels in thermal spas and the compliance with the European BSS directive: a Portuguese case study. In: Occupational and Environmental Safety and Health II. Springer, Cham, pp 163–171

Soto J, Gómez J (1999) Occupational doses from radon in Spanish spas. Health Phys 76:398–401

Soto J, Fernandez PL, Quindos LS, Gómez-Arozamena J (1995) Radioactivity in Spanish spas. Sci Total Environ 162(2–3):187–192

Stojanovska Z, Boev B, Zunic ZS, Ivanova KG, Ristova M, Tsenova M, Ajka S, Janevik E, Taleski V, Bossew P (2016) Variation of indoor radon concentration and ambient dose equivalent rate in different outdoor and indoor environments. Radiat Environ Biophys 55(2):171–183

Stojanovska ZA, Ivanova K, Bossew P, Boev B, Žunić ZS, Tsenova M, Surguz Z, Kolar P, Zdravkovska M, Ristova M (2017) Prediction of long-term indoor radon concentration based on short-term measurements. Nuclear Technol Radiat Prot 32(1):77–84

Szerbin P (1996) Natural radioactivity of certain spas and caves in Hungary. Environ Int 22:389–398

Turtiainen T, Kojo K, Laine JP, Holmgren O, Kurttio P (2021) Improving the assessment of occupational exposure to radon in aboveground workplaces. Radiation Protection Dosimetry

UNSCEAR (2000) Sources and effects of ionising Radiation, Volume 1. Report to the general assembly with scientific annexes. Annex B. Exposures from natural radiation sources. UNSCEAR 2000 Report. United Nations: New York

Vassileva S (1996) Mineral water and spas in Bulgaria. Clin Dermatol 14(6):601–605

Vaupoti J, Kobal I (2001) Radon exposure in Slovenia spas. Radiat Prot Dosim 97(3):265–270

Vinson DS, Campbell TR, Vengosh A (2008) Radon transfer from groundwater used in showers to indoor air. Appl Geochem 23(9):2676–2685

Vogiannis E, Nikolopoulos D, Louizi A, Halvadakis CP (2004) Radon variations during treatment in thermal spas of Lesvos Island (Greece). J Environ Radioact 75(2):159–170

WHO (2009) In: Handbook on indoor radon: a public health perspective. In: Zeeb H, Shannoun F (eds) World Health Organization

Žunić ZS, Kobal I, Vaupotič J, Kozak K, Mazur J, Birovljev A, Janik M, Celiković I, Ujić P, Demajo A, Krstić G, Jakupi B, Quarto M, Bochicchio F (2006) High natural radiation exposure in radon spa areas: a detailed field investigation in Niška Banja (Balkan region). J Environ Radioact 89(3):249–260