

# Analysis of Exposure To Radon in Bulgarian Rehabilitation Hospitals

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## Research Article

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52 The Council of the European Union issued the European Basic Safety Standards (EU BSS) in December.  
 53 2013, which recommended that the annual average radon concentration should not be higher than 300  
 54 Bq/m<sup>3</sup> in dwellings, workplaces and building with public access (EC, 2013).  
 55 Bulgaria has an abundance of mineral water springs located throughout its territory (Hristov et. al, 2016).  
 56 According to Vassileva, (1996), the number of mineral springs in Bulgaria is over 520, with different  
 57 compositions, temperatures, and properties of mineral water with most of them being warm. 57 of the  
 58 230 existing spas are promoted as balneological resorts (Hristov at. al., 2019). There have been certain  
 59 investigations of natural radioactivity and radon in mineral water in the literature, which shows the  
 60 presence of a high concentration (Kamenova-Totzeva et. al., 2018). This implies that in spa or in  
 61 rehabilitation centres, staff and patients may have been exposed to higher levels of radon, leading to  
 62 significant additional exposure. Although Bulgaria has adapted its radiation protection standards based  
 63 on the European directive, there exists no systematic indoor radon measurements in public buildings  
 64 where mineral water is used.  
 65 This paper focused on the results of indoor radon ( $C_{Rn}$ ) and radon in water ( $C_{Rnw}$ ) measurements obtained  
 66 via a survey of 12 thermal specialized hospitals for rehabilitation in Bulgaria. The aim of the survey,  
 67 realized in the framework of a project funded by Bulgarian National Science Fund, was to investigate  
 68 radon levels and perform a radon exposure assessment in buildings with public access. Thus, the national  
 69 measurement protocol for indoor radon in buildings with public access, where the thermal water is used,  
 70 was verified.

## 71 2. Materials and method

### 72 2.1. Objects and design of survey

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

82 All the branches (11) of the national complex are located in mountainous regions, except for one present  
 83 in the Danube plain (the village of Ovcha Mogila). The locations within the territory of Bulgaria are  
 84 presented in Figure 1. The specialized hospitals are located primarily in small spa resorts, with only  
 85 Kyustendil being a relatively large town and the administrative centre of the district. Five of these  
 86 specialized hospitals are situated within one building, while the rest are in two or three buildings. The  
 87 bathrooms with pools in the branch at Banya-Karlovo are situated in two separate small buildings on  
 88 one floor. Further, an additional building of the Ministry of Health was investigated in Velingrad, which  
 89 was also used for rehabilitation purposes. The exact number of buildings are presented in Table 1. The  
 90 buildings are usually large having several floors (from 2 in Pavel Banya and Varshets to 9 in Narechen)  
 91 and the total area of these buildings varies from 360 to 8500 m<sup>2</sup>. The underground floors in the hospitals  
 92 are usually occupied with, a swimming pool or treatment rooms situated in them, but in some of  
 93 hospitals, the underground floor is used for storage of mineral water in tanks or for warehouses and  
 94 workshops.

95  
 96 *Table 1. Location of surveyed specialized rehabilitation hospitals with their code, number of*  
 97 *buildings, measured premises with and without using water for the treatment. Percentage of losses of*  
 98 *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, Kosteneec, 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
<b>Total</b>		<b>21</b>	<b>291</b>	<b>110</b>	<b>18</b>

99

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



105

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

107 The indoor radon ( $C_{Rn}$ ) in treatment rooms, pools and offices on the basement and ground floor were  
 108 measured with passive detectors. Measurements were also performed on the first, second and upper  
 109 floors, but not in all premises. Radon CR-39 detectors in premises without water procedure were  
 110 positioned on a shelf at a distance of approximately 1 – 1.5 m above the floor or any wall surface. In  
 111 premises where water is used, such as swimming pools and bathrooms, the detectors were suspended  
 112 via a rope from the ceiling or mounted on partitions separating showers, bathtubs, or treatment rooms.  
 113 For the measurements of radon concentration in water ( $C_{Rnw}$ ), samples were procured from the spring  
 114 (borehole or casing) and from the treatment rooms (baths). The number of detectors, being divided into  
 115 rooms in which water is used for treatment and ordinary premises (offices, doctors' offices, rooms) are  
 116 presented in Table 1. In the rehabilitation hospitals, as with public facilities, there are losses of detectors

117 because they are visited by many people, and at the end of the exposure period, several of the detectors  
 118 were missing. Higher losses were expected because the surveyed buildings are used by a lot of peoples.  
 119 The total losses amounted to 18 %, and the distribution by branches is summarised in Table 1. The  
 120 biggest losses were faced at the branches of Banya, Karlovo and Velingrad. The bathrooms, in these  
 121 branches, are present in separate buildings and are visited by more people, which could explain the  
 122 losses. Thus, the total number of analysing detectors were 401, with 291 detectors on premises where  
 123 water was not used for treatment and 110 in premises with water procedures present, such as pools,  
 124 baths, procedure rooms.

## 125 2.2. Indoor radon measurement

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

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

$$144 C_{Rn} = F_c \cdot (d - b) \cdot t^{-1} \quad (1)$$

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

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

$$148 u() = \sqrt{\left(\frac{u(F_c)}{F_c}\right)^2 + \left(\frac{u(d)^2 + u(b)^2}{(\bar{d} - \bar{b})^2}\right) + \left(\frac{u(t)}{t}\right)^2} \quad (2)$$

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

## 160 2.3. Sampling and measurement of radon in water

161 The method based on Passive Environmental Radon Monitor (E-PERM) Electret Ion Chamber (EIC)  
 162 system manufactured by Rad Elec Inc. was used for the measurement of the radon concentration in  
 163 water, which consists of a reader, S-chambers and long (LT) or short –term (ST) electret, jars of 3.72 l  
 164 volume and sample bottles of 68 and 136 ml. The water samples were collected using 136 ml collection  
 165 bottles. Further, a bucket was used to till it overflowed, after the water ran for several minutes, and  
 166 thereafter, raised such that the tap (or source) was below the surface. The sampling bottle was  
 167 subsequently submerged and filled from the bottom of the bucket and capped underwater (Kotrappa,  
 168 1999). Three parallel bottles from the sampling point were procured. Two of them were measured  
 169 simultaneously, while the third was a control sample and measured when the results of the first two

170 differed. The notes for exact data and hour, ID of sample and duration of transport to laboratory were  
 171 maintained. In the laboratory the sampling bottle was placed without cap at the bottom of the glass jar,  
 172 and an E-PERM short-term chamber with a long-term electret or configuration of ionizing chamber - SLT  
 173 was suspended over the water spread on the bottom. Subsequently, the jar was sealed and remained  
 174 closed for 24-48 h and the exact duration of period was written in protocol of the sample. The evaluation  
 175 of radon concentration in water ( $C_{RnW}$ ) was based on radon concentration in air inside jar  $C_{Rnj}$ ,  
 176 normalized to the volume of water sample and the jar and two periods. ( $T_d$  - the period utilized for  
 177 collection of samples till immediately before insertion of the sample for analysis and  $T_a$  - the period  
 178 utilized for inserting the sampling bottle into the measuring jar till the ionizing chamber is removed for  
 179 analysis) The measurement of  $C_{Rnj}$  inside the jar was estimated under the voltage difference before and  
 180 after exposure of detector in jar, gamma dose rate in laboratory and appropriate calibration factors, based  
 181 on the procedure described by the producer (Rad. Elec. Inc., Frederick, Maryland, USA). Therefore, the  
 182  $C_{RnW}$  was calculated after measuring the  $C_{Rnj}$  by multiplying by the constants  $C_1$ ,  $C_2$  and  $C_3$  as follows

$$183 \quad C_{RnW} = C_{Rnj} \cdot C_1 \cdot C_2 \cdot C_3 \cdot \quad (3)$$

184 where, the constant  $C_1$  considers the delay period between the collection of the water sample and the  
 185 beginning of the measurement and is expressed as

$$186 \quad C_1 = e^{(\lambda \cdot T_d)}, \text{ as } \lambda=0.1814 \quad (4)$$

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

$$188 \quad C_2 = \frac{\lambda \cdot T_a}{1 - e^{(-\lambda \cdot T_a)}} \quad (5)$$

189 The constant  $C_3$ , is the ratio between the volume of the jar and that of the sampling bottle, and this case  
 190 was equal to 28. In addition, the reader was regularly sent to the producer for calibration. The method  
 191 was validated with the encapsulated  $^{226}\text{Ra}$  source in water during the intercomparison (Kitto, et al.,  
 192 2010) and the estimated combined uncertainties of the method was 20 % (at 95 % confidence level).

#### 193 *2.4. Data analysis*

194 The results obtained were evaluated and systematized, and the information from the completed  
 195 questionnaires was summarised for analysis. This was achieved by employing the SPSS (version 19)  
 196 statistical software. In the analysis, depending on the distribution and homogeneity of the grouped  $C_{Rn}$   
 197 results, appropriate parametric and non-parametric statistical tests were used at a 95 % confidence level.  
 198 Based on the data, the average transfer factor was set as the ratio of the radon concentration in the air to  
 199 that in water in the treatment premises ( $f_i = C_{Rn}/C_{RnW}$ ), which describes the transfer of radon from water  
 200 to air. Although the transfer factor depends on the water temperature (Cosma et al., 2008), in this study  
 201 the influence of temperature was neglected because the water temperature in the pools and baths of the  
 202 hospitals were maintained at approximately the same temperature (approximately 30 °C). Depending on  
 203 the temperature of the water in the springs, it is heated or cooled before being introduced into the baths.  
 204 To assess the radon exposure of patients and workers in rehabilitation hospitals the annual effective dose  
 205 ( $E$ ) was estimated according the EU Radiation Protection N° 193 (2020) as follows

$$206 \quad E = C_{Rn} \cdot F \cdot t \cdot C_f \quad (6),$$

207 where  $E$  is expressed in mSv,  $C_{Rn}$  is the average indoor radon concentration in  $\text{Bq/m}^3$  in premises with  
 208 water procedure and  $F$  is the equilibrium factor between radon gas and its decay product. The value of  
 209 the equilibrium factor,  $F$ , depends primarily on the indoor ventilation rate because of opening/shutting  
 210 of windows, and the use of electric fans and air conditioners. Although the equilibrium factor in thermal  
 211 spas, according to some authors varies from 0.2 (Vogiannis et al., 2004) to 0.6 (Soto and Gomez, 1999),  
 212 the standard assumption of  $F = 0.4$  was applied for calculation (UNSCEAR 2000; ICRP, 2017). Further,  
 213  $t$  is exposure time in hours. Effective doses of inhalation of radon in rehabilitation hospitals by a  
 214 reference worker and patient were calculated assuming the breathing rate of  $1.2 \text{ m}^3 \text{ h}^{-1}$ , that is,  
 215 approximately one-third of time spent sitting and two-thirds of time spent in light exercise. In addition,  
 216  
 217

218 for the calculation of radon effective doses, a dose coefficient of 3 mSv per mJ h m<sup>-3</sup> (approximately 10  
 219 mSv/WLM) corresponding to  $C_f = 6.7 \cdot 10^{-6}$  mSv/[(Bq.h/m<sup>3</sup>)] was applied (ICRP, 2017). The dose  
 220 coefficients were calculated using defined biokinetic and dosimetric models for reference person under  
 221 particular exposure conditions as reference values and were not regarded as subject to uncertainty  
 222 (ICRP, 2007). The sources of uncertainties in biokinetic models are associated with the types of  
 223 information used to construct the models for the elements. Furthermore, the uncertainty in the dose  
 224 assessment depends on uncertainties associated with measurements of radon concentration and sampling  
 225 and uncertainties in the exposure scenario, including factors such as period of exposure. However, for  
 226 regulatory purposes, the models and parameter values were fixed by convention.

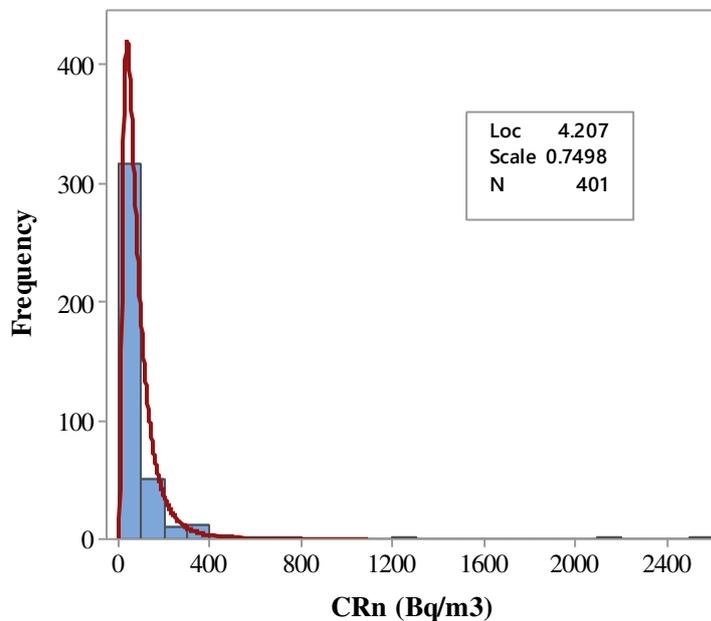
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### 228 3. Results and discussion

#### 229 3.1. Radon concentration in air

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

235



236

237 *Figure 2. Histogram of  $C_{Rn}$  fitted with log normal function*

238

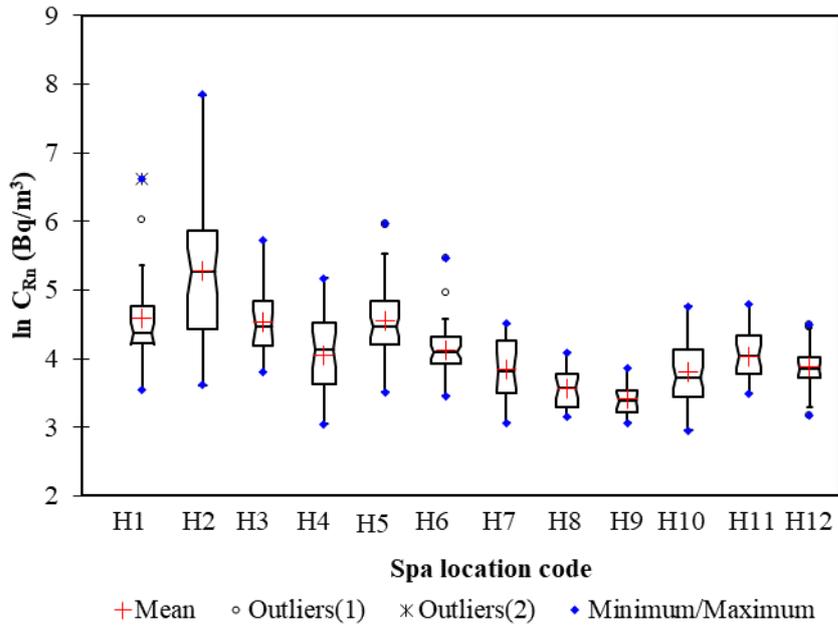
239 To evaluate the spa region, the measured buildings were grouped by location and Table 2 presents the  
 240 descriptive statistic and  $p$ -value of Shapiro–Wilk test for normality of  $\ln C_{Rn}$  distribution ( $p > 0.05$ , 95 %  
 241 confidence level). Considering the  $p$  values in Table 2, it follows that the ln-transformed results from  
 242 most spa regions follow a normal distribution or the data are in the vicinity the mean value more often.  
 243 However, the data of Narechenski Bani (H1), Pavel Banya (H5), Varshets (H6) and Velingrad (H10)  
 244 spa regions do not exhibit a normal distribution. However, to avoid the influence of extreme values all  
 245 further analyses were performed on  $\ln C_{Rn}$ . The reasons for inhomogeneity of the  $C_{Rn}$  can be many. The  
 246 results of  $C_{Rn}$  are influenced by differences in geology between locations but also differences in  
 247 premises. As a result, there is a wide range of  $C_{Rn}$  values between the locations and on the locations  
 248 themselves. The measured  $C_{Rn}$  have a minimum value of 19 Bq/m<sup>3</sup> in the Velingrad branch (H10) up to  
 249 2550 Bq/m<sup>3</sup> in . The significant difference between  $C_{Rn}$  in the different spa regions was confirmed by  
 250 the Kruskal-Wallis test (KW,  $p < 0.0001$ ). Furthermore, we tested the difference between  $C_{Rn}$  in each  
 251 two regions separately via the Mann-Whitney test, which conforms to the assumption of their grouping  
 252 Fig. 3. We find all differences are significant with  $p < 0.05$  between the locations:  
 253 H2 > H1, H3, H5 > H4, H6, H11 > H7, H10, H12 > H8, H9

254 The highest  $C_{Rn}$  was found at Momin Prohod (H2), which implies that it is a spa region with high radon.  
 255 The variation coefficient was high too, indicating the high range of  $C_{Rn}$ . The average  $C_{Rn}$  of measured  
 256 buildings in Momin Prohod were found higher than reference level of  $300 \text{ Bq/m}^3$ . In a similar manner,  
 257  $C_{Rn}$  values were found to be high for the region of Niska Banja, Serbia by Žunić, et. al., (2006). In  
 258 addition, the results of a survey conducted in Spain show a  $C_{Rn}$  of over  $5000 \text{ Bq/m}^3$  (Soto, J, et. al.,  
 259 1995), which were higher than the results in Momin Prohod. Further, Narechen (H1), Banya-Karlovo  
 260 (H3) and Pavel Banya (H5) were found to be the spa regions with moderate  $C_{Rn}$  values. These regions  
 261 need to be further investigated in detail, and actions are required to inform the public in accordance with  
 262 the national radon plan. The arithmetic mean value of  $C_{Rn}$  in the region with low radon in the range from  
 263  $30,9 \text{ Bq/m}^3$  in Sandanski (H9) to  $37 \text{ Bq/m}^3$  in (H8). The coefficient of variation, which depicts the  
 264 extent of variability in relation to the mean of the data in those regions, was also relatively small. The  
 265 results from low radon spa region are in the similar variation range, as in the thermal baths of Rimini  
 266 and Pesaro-Urbino provinces, Central Eastern Italy ( $7\text{-}71 \text{ Bq/m}^3$ ), published by Desideri et.al (2004).  
 267 Moreover, the  $C_{Rn}$  in five Slovenian spas, at Rogaska Slatina, Radenci, Moravci, Podcetrtek, and Catez  
 268 were in the same low range, because of the effective ventilation systems (Vaupoti and Kobal, 2001).  
 269

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

Code of location	Indoor radon concentration ( $C_{Rn}$ )							
	N	AM, $\text{Bq/m}^3$	SDV, $\text{Bq/m}^3$	Min, $\text{Bq/m}^3$	Max, $\text{Bq/m}^3$	CV, %	GM, $\text{Bq/m}^3$	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

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

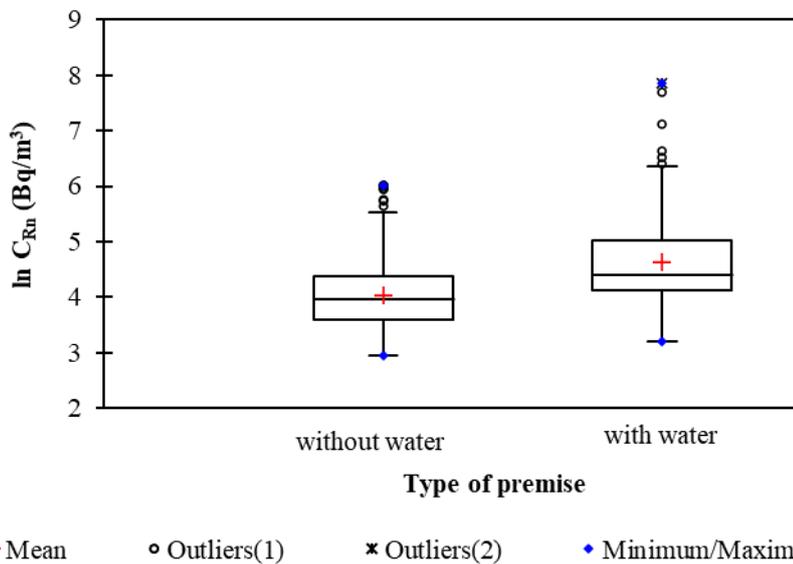


274

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

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

279

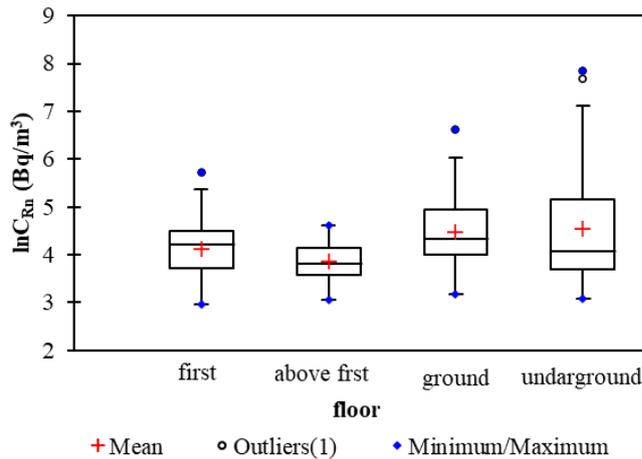


280

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

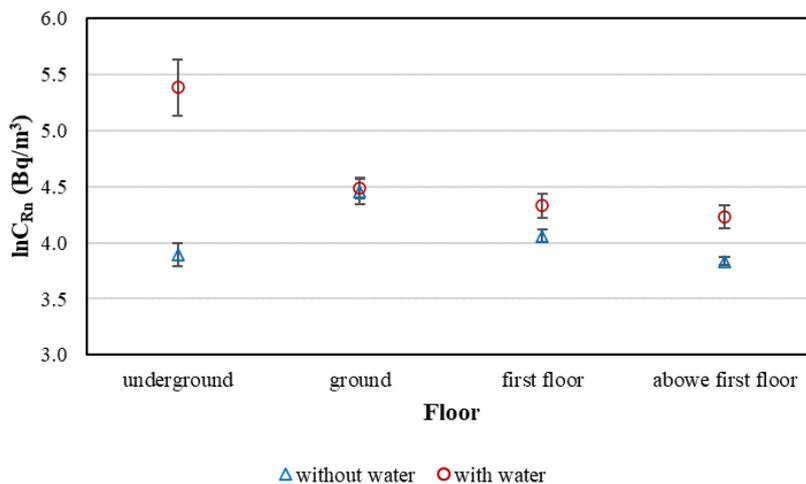
283 The arithmetic mean value for the procedure rooms was calculated to be  $AM = 186 \text{ Bq/m}^3$ , which was  
 284 approximately three times higher than in rooms without water procedures ( $AM = 77 \text{ Bq/m}^3$ ).  
 285 Subsequently, a non-parametric Mann-Whitney test was applied for the two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

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



290  
291 *Fig. 5 The boxplots of  $\ln C_{Rn}$  by floors in the surveyed specialized rehabilitation hospitals*

292 Subsequently, a non-parametric Kruskal-Wallis test was applied and a statistically significant difference  
 293 between the groups (KW,  $p < 0.001$ ) was confirmed. To further check between which groups a difference  
 294 existed, the Mann-Whitney rank test was used for all couples and no statistically significant difference  
 295 was observed between the underground, ground and first floor. However, a difference was found  
 296 between the group with results above the first floor and the lower floors (MW,  $p < 0.001$ ) which can be  
 297 explained by the location of most treatment premises on the lower floors. This finding suggest that water  
 298 could be an significant source of radon in most premises. It can be noticed from Fig. 6, the  $C_{Rn}$  in the  
 299 underground rooms without water treatment are much lower than in the treatment rooms where the water  
 300 further increases the  $C_{Rn}$ . Given that the most treatment rooms are located on the ground floor, we  
 301 assume that radon from the water further increases the indoor concentration in all rooms. On the first  
 302 and upper floors, the effect of geogenic radon weakens and the  $C_{Rn}$  decreases in rooms without  
 303 treatments. In the treatment rooms on those floors, the  $C_{Rn}$  is almost the same, indicating that water is  
 304 an additional source of indoor radon.



305  
306 *Fig. 6 The mean values of  $\ln C_{Rn}$  grouped by the type of premise and floor*

### 307 3.2. Radon concentration in water

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

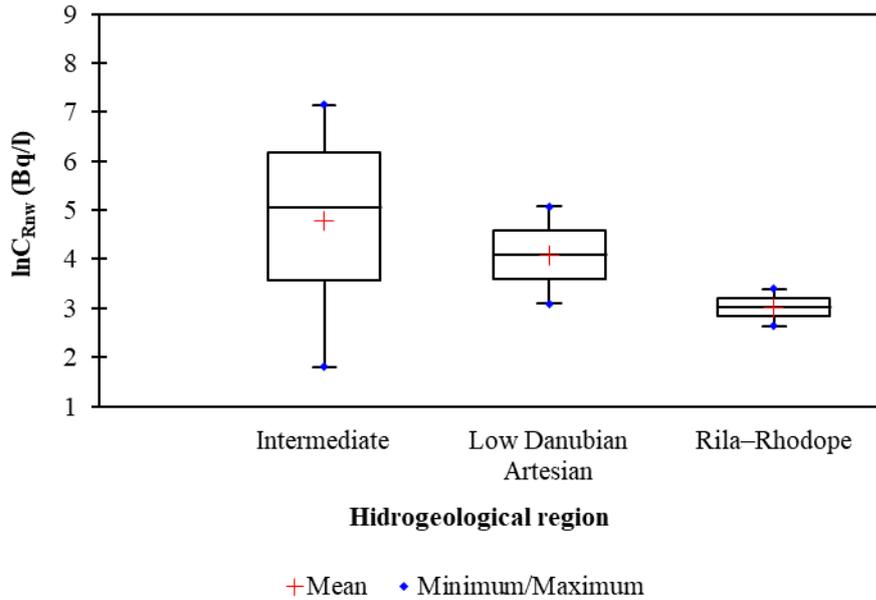
314 In the rehabilitation hospitals at Banya, Karlovo (H3) and Velingrad (H10), thermal water is used  
 315 directly in the bathroom from the spring. The  $C_{RnW}$  from the springs at Narechenski Bani (H1), Momin  
 316 Prohod (H2), Pavel Banya (H5) and Velingrad (H10) were above the standards set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

325 *Table 3. Radon concentrations in springs and bath water, average radon concentration in bath air*  
 326 *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 <sup>3</sup> (treatment premises)	Transfer coefficient
Narechenski Bani(H1)	28	613	189	186	$9.8 \cdot 10^{-3}$
Momin Prohod (H2)	64.5	1265	270	795	$2.9 \cdot 10^{-3}$
Banya, Karlovo (H3) - men's bath	25	19		125	$6.6 \cdot 10^{-3}$
Banya, Karlovo (H3) - women's bath	26	17		138	$8.1 \cdot 10^{-3}$
Banya, Karlovo (H3) –central spring	34,5	58		166	$2.9 \cdot 10^{-3}$
Hissarya (H4)	45	30	15	62	$4.1 \cdot 10^{-3}$
Pavel Banya (H5)	61	428	223	95	$4.3 \cdot 10^{-4}$
Varshets (H6)	37,6	22	31	72	$2.32 \cdot 10^{-3}$
Bankya (H7)	23	24	4.3	36	$8.37 \cdot 10^{-3}$
Kyustendil (H8)	73	6	11	45	$4.02 \cdot 10^{-3}$
Sandanski (H9)	72	30	23	28	$1.21 \cdot 10^{-3}$
Velingrad (H10) – Kamena spring	47	222		86	$3.87 \cdot 10^{-4}$
Velingrad (H10) – Veliova bania spring	63	110		135	$1.23 \cdot 10^{-3}$
Ovcha mogila (H11)	45	159	32	77	$2.41 \cdot 10^{-3}$
Banite, Smolian (H12)	42	14	16	78	$4.88 \cdot 10^{-3}$

327 Spearman's rank correlation coefficient was used to assess the relationship between the concentration of  
 328 radon in mineral water and in indoor air. The test is statistically significant ( $p < 0.05$  at 95% confidence  
 329 interval) and Spearman's correlation coefficient between the  $C_{RnW}$  from baths and  $C_{Rn}$  was  $\rho_o = 0.806$ .  
 330 The correlation analysis showed that 65 % ( $\rho^2 = 0.65$ ) of radon measurements in air can be explained by  
 331 the presence of radon in bath water, that is, high values of radon in the air could be expected to be found  
 332 if there are high values of radon in the water. To systematize the results and link them to hydrogeology,  
 333 the  $C_{RnW}$  results from the springs were grouped by the hydrogeological regions of the country. Bulgaria  
 334 has been divided into three major hydrogeological regions, which is closely linked to the main geological  
 335 structures, namely, low Danubian Artesian, intermediate, and Rila–Rhodope (Benderev, et. al., 2016;  
 336 Hristov, et. al., 2019).

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



344  
345 *Fig. 7 The boxplots of  $\ln C_{RnW}$  from springs by hydrogeological regions in Bulgaria*

346  
347 *3.3. Estimation of exposure to radon*

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

353  
354  
355  
356 *Table 4. Estimated effective doses from radon inhalation by specialized hospital for rehabilitation of*  
357 *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
Velinograd (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

359 The main uncertainty in calculation of effective dose was the exposure period, which was applied. The  
360 working hours in the premises with water therapy, such as, swimming pools, bathtubs, tangents, and  
361 others have been reduced and are 1750 hours per year. In contrast, at the other workplaces in the  
362 rehabilitation hospitals the working hours are 2000 hours a year. To calculate the additional annual  
363 effective dose ( $E$ ,  $\mu\text{Sv}/\text{y}$ ) of a patient receiving a full treatment programme, it was considered that each  
364 patient receives 30 treatments annually (one month) of 1-hour duration in the pool, bath, or other  
365 premises with water. The estimated effective doses from radon inhalation by specialized hospitals for  
366 rehabilitation of workers and patients are presented in Table 4. The effective doses because of radon  
367 inhalation of workers in premises with water were higher and varied from 0.3 to 14.4  $\text{mSv}/\text{y}$  depending  
368 to radon concentration in rehabilitation hospitals. As expected, the workers in the rehabilitation hospital  
369 at Momin Prohod received the highest dose. The effective doses of the workers were in range of the  
370 assessed doses of workers in the thermal spas of Ischia Island (Pugliese, et, al., 2014). In contrast, the  
371 estimated effective doses of radon inhalation for patients are relatively low and range from 2.0 to  
372 250  $\mu\text{Sv}/\text{year}$ . Though these doses do not exceed the limit of the annual effective dose for the population  
373 from all sources (1  $\text{mSv}/\text{year}$ ), in some regions (Narechenski Bani and Momin Prohod) they were higher  
374 than the levels of exemption of 10  $\mu\text{Sv}/\text{year}$  according to the Bulgarian legislation standards.

#### 375 **4. Conclusion**

376 Radon measurements in the air and geothermal water of 12 Bulgarian rehabilitation hospitals were  
377 performed and the indoor radon concentrations and water of the springs and baths were obtained in the  
378 range of 19 to 2550  $\text{Bq}/\text{m}^3$ , 6 to 1265  $\text{Bq}/\text{l}$  and 4.3 to 270  $\text{Bq}/\text{l}$ , respectively. The correlation analysis  
379 was used to test relationships between the data of the concentration of radon in the mineral water and in  
380 the air in the treatment rooms and the Spearman's rank correlation coefficient was calculated to be  
381  $\rho=0.806$ , which proves that the connection of the levels of indoor radon is related to those of the water.  
382 Further, the difference in radon concentration in the premises with water therapy and those without was  
383 confirmed. The results clearly show that thermal water with high radon concentration is the source of  
384 radon in buildings. In addition, the analysis and assessment of exposure to radon confirmed the need of  
385 radon control in spas to adhere to the European Basic Safety Standard (EU BSS, Council Directive  
386 2013/59/Euratom, 2014), which could be realized by inspecting the ventilation system, an important  
387 part for improving the indoor environment in spa. The five spa regions were identified according the  
388 indoor radon concentration which could be used to apply a graded approach in the control of spas.  
389 Further, an analysis of the results of radon concentration in water from springs by their hydrogeological  
390 regions was performed. Intermediate hydrogeological region had the lowest arithmetic means of the  
391  $C_{\text{RnW}}$  from springs ( $C_{\text{RnW}}=25.7 \text{ Bq}/\text{l}$ ), while Rila–Rhodope region had the highest ( $C_{\text{RnW}}=375.5 \text{ Bq}/\text{l}$ ).  
392 Further detailed study of the geology of spa areas should be performed for assessment of the factors that  
393 influence the radon concentration in water. Finally, the estimation of annual effective doses for workers  
394 and patients were evaluated considering the exposure period and the measured indoor radon  
395 concentration. Although the study was done in only 12 regions with mineral water in the specialized  
396 rehabilitation hospitals, the results concerning the indoor radon and water variation could be used to  
397 optimize future radon surveys in buildings with public access as spa centres.

#### 398 **Ethical Approval and Consent to Participate**

399 We declare that this manuscript is original, has not been published before and is not currently being  
400 considered for publication elsewhere. No data, text, or theories by others authors are presented, except  
401 the citations on manuscript. The results are measured and analysed by authors of the manuscript.

402 Our manuscript not involving human participants, human data or human tissue and animals.

#### 403 **Consent to Publish**

404 Not applicable

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#### 408 **Authors Contributions**

409 All authors contributed to the study conception and design. Material preparation, data collection and  
410 analysis were performed by Desislava Djuvnačkova, Kremena Ivanova and Bistra Kunovska. Statistical  
411 analysis was done by Zdenka Stojanovska. The first draft of the manuscript was written by Jana Jounova  
412 and Nina Chobanova and all authors commented on previous versions of the manuscript. All authors  
413 read and approved the final manuscript. Corresponding author is Kremena Ivanova.

#### 414 **Competing Interests**

415 The authors declare that they have no competing interests

#### 416 **Availability of data and materials**

417 The datasets generated and analysed during the current study are available in the NCRRP data base and  
418 are available from the corresponding author on reasonable request. The data report are publicly available  
419 on the [www.radon.bg](http://www.radon.bg)

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