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RADON IN WATER FROM PRIVATE WELLS AND ITS CONTRIBUTION TO INTERNAL EXPOSURE OF POPULATION IN RURAL AREAS AT TOPLICA REGION, SOUTHERN SERBIA

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ABSTRACT

The results presented in this paper are part of the investigating of radon the concentrations from natural sources in the Toplica region. The results refer only to the radon concentrations in water from private captured wells at 12 locations. Radon concentration in water was measured by alpha spectrometry using the RAD 7 – RAD H2O system. The range of radon concentration is from (2.8±1.2) to (76.0±4.0) kBq/m^3, and the contribution of radon released from water to the air in the premises was in range (0.8±0.3) to (22.8±1.2) Bq/m^3. The annual effective doses of inhaled and ingested radon were determined, the mean values were (114.8±14.8) and (3.2±0.3) μSv/y.

Keywords: Radon in water, Private wells, Annual effective radiation doses.

INTRODUCTION

Groundwater is the largest freshwater resource and it is very important to investigate radon in groundwater to protect the population from health hazards caused by radon and its progeny (UNSCEAR 2000). Radon is a radioactive gas product of the uranium sequence and a direct progeny of radium. It is characterized by a half-life of 3.82 days, four short-lived progeny and is an important source of alpha particle emission (Ravikumar et al., 2014; Di Carlo et al., 2019). In the literature, radon from water is recognized as secondary sources of indoor radiation (WHO 2004; Anjos et al., 2010; Rožmarić et al., 2012; Kendall & Smith 2002; Isinkaie et al., 2021). Radon is soluble in water, leaves it very quickly under pressure or at elevated temperature and accumulates in the air in a closed space and thus can cause damage to organs, tissues and cause changes in the DNA chain (Fakhri et al., 2016). Radon in water can lead to body irradiation in two ways: by ingesting radon-rich water and by inhaling radon that is released from it into the interior of the room when the water is used for bathing, cooking, washing, etc. (UNSCEAR 2000). Radon moves from the water into the closed space through the process of diffusion, so it can be said that the concentration of radon in the closed space increases simultaneously with the high content of radon in the water.

Although the radiation dose from radon ingestion from drinking water is lower than the radiation dose from inhalation, excessive radon intake from drinking water can not only cause lung cancer, but can also increase the risk of stomach and colon cancer (Bonotto 2004; WHO 2004; Anjos et al., 2010; ICRP 2010; Vogeltanz-Holm & Schvartz, 2018).

Population in Toplica region is supplied with drinking water from natural sources of underground water in the selected localities. The aim of these studies was to determine the radon concentration in water from private captured wells, to calculate the contribution of radon from water to the total radon in the air in a closed space, and to assess to what extent is the population exposed to radiation through ingestion and inhalation of radon.

STUDY AREA

Toplica region is a medium-developed industrial center and an important fruit growing and agricultural area of Serbia. Toplica region situated in the South Serbia with geographical coordinates 42°52′ – 43°24′N of latitude and 20°56′ – 21°50′E of longitude (Fig.1). The area of Toplica region is 2180 km^2. According to the last census in 2022, the total number of inhabitants is 77.900 (SORS, 2023), making it the smallest region by population in Serbia.

This geographical area is defined by the basins of the Toplica and Kosanica rivers. This sub-region is bordered in the east with the South Morava river, in the west with the Mountain Kopaonik. The south border occupied territory with the Mountains Majdan and Radan. The border in the north of the Toplica region are Mountain Veliki and Mali Jastrebac (Pavlović, 2019). The administrative regionalization of Toplica followed four municipalities: Prokuplje, Kuršumlija, Žitorađa and Blace.

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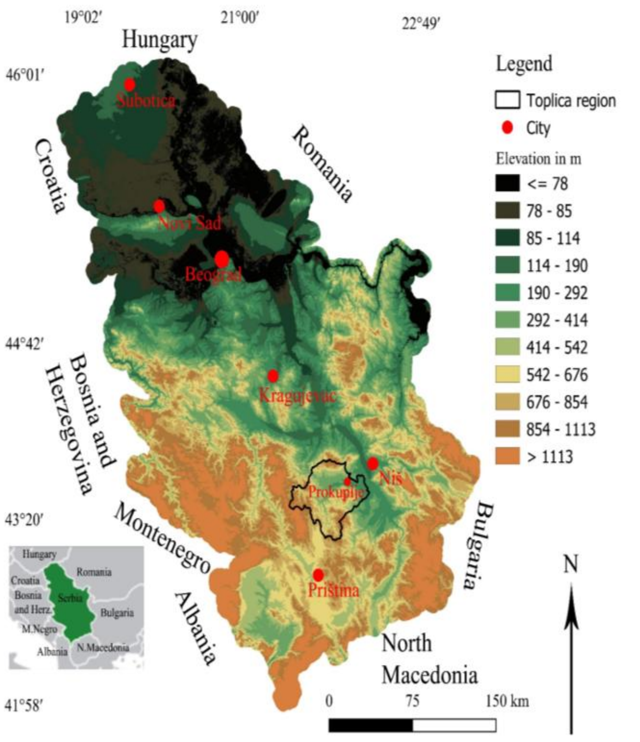

Figure 1. The geographical position of Toplica region in the Republic of Serbia.

MATERIAL AND METHODS

The paper presents the results of radon concentration of radon in water that the population uses both for drinking and for other purposes from 12 selected localities in the Toplica region. In this research seven samples were taken in Kuršumlija, one in Blace, one in Žitorađa and three in Prokuplje (Fig.2).

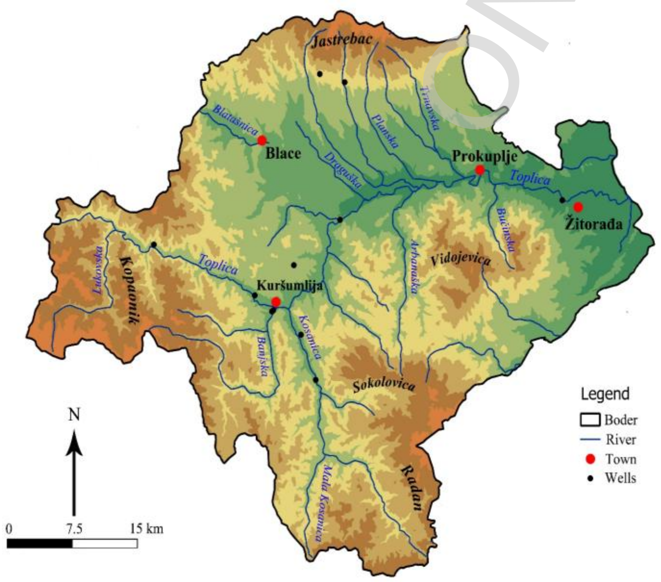

Figure 2. The position of private wells in the Toplica region.

GIS and remote sensing methodology

In this research the GIS methodology were used to analyze better spatial distribution of wells. The wells distributions are approved by handled Garmin GPS 72H with an accuracy of 3 m. All data were used and analyzed with this device and after that mapped. The geodetic projection was WGS 84 4326 and this projection is reprojected to be at national scale and national coordinate system. The national projection is UTM zone 7. This projection is good because is in the correlation with old topographic maps (Valjarević et al., 2018). GIS methods, such as the method of interpolation and graduated method are provided to present the final geographical position (latitude and longitude) of analyzed wells (Fig. 2). These methods also showed very clearly the position of wells.

Geological structure of studying area

In the geological past the Toplica region was a region with very strong volcanic activity (Geological Atlas of Serbia 2002). The Rhodopes are considered as the oldest mountains of the Balkan Peninsula and Serbia; they are built of the archaic and Paleozoic crystalline schist (Stevanović et al., 2018). Even today in this region there are many paleovulcanic forms. The main types of crystalline schists are andesite, fine-grained gneisses, amphibolites, magmatites, leptinoliths, mica schists, quartzite, marble, amphibole schist, pegmatite and mica rocks. Due to volcanic activity the most dominant rocks are andesite. Green shale and metamorphosed gabbro found on the mountain Jastrebac belong to the Cambrian rocks, while low metamorphosed rocks from Devon period have been discovered in tectonic contacts of crystalline shale, serpentinisedperidotite and Senonian sediments (Geological Atlas of Serbia 2002; Valjarević et al., 2014). From the Mesozoic era, the oldest rocks are related to Middle Triassic and widespread northwest of Kuršumlija. The rocks formed during the Late Jurassic are positioned in the west of the region in the form of mass or elongated, but discontinuous zones having the direction of the NNW–SSE are presented by basic and ultra-basicmetamorphites and diabase-chert formation (Geological Atlas of Serbia 2002). This geological structure supported occurrences of thermo-mineral springs and underground wells with high concentration of radon. Observing the geomorphological structure of the terrain, the examined terrain contains various rock masses subjected to physical and chemical changes in the surface parts. It allows water to accumulate in the systems of cracks and fissures, the degree of which is variable and depends on the external conditions. A detailed description of geological structure of chosen private wells is given in Table 1. which based on the detailed geological map of Toplica region.

Population is supplied with drinking water from natural sources, due to unreliable supply from the public water supply system. The sources from which the population is supplied with drinking water are captivated natural wells in the immediate vicinity of the houses, so that the spring water is carried inside the houses through a closed system of pipes. There is a fountain in every yard at the point where the spring is. Family houses built in the midterm of 20th century, with concrete floors, without basement rooms and approximately similar layout of rooms were selected. Rooms where inhabitants spent the most of the time are the living rooms, the kitchen is most often an integral part of them, and they are of the same or very similar dimensions.

Table 1. The geological structure of chosen private wells.

No	Geological background
1	Proluvium
2	Coarse-grained sandstones and microconglomerates
3	Pleistocene river terrace of the Banjska river
4	Jurassic diabase and spilite
5	Pleistocene river terrace of the Banjska river
6	Senonian siltstones, marlstones and thinly-layered limestones
7	Volcanic breccias and tuffs
8	Red series. siltstones and sandstones
9	Proluvial deluvial sediments
10	Marbles
11	Alluvium
12	Proluvium

Sempling procedure and detection of radon

Before sampling, the water temperature was measured with a digital thermometer (Testo Se & KGaA, Germany). After that, the water was sampled in original glass bottles with a volume of 250 ml up to the very top. When filling the vials, water flowed from the tap in a small stream, in order to avoid the formation of bubbles in the sample. The closure was performed under a jet of water, which prevented the forming of a free air space under the closure, in which radon could accumulate due to exhalation from the water. Since the concentration could not be measured during the sampling itself, it was necessary to deliver the samples as soon as possible to the Laboratory for testing the radioactivity of samples and doses of ionizing and non-ionizing radiation at the Faculty of Science, University of Novi Sad, where the radon concentration in water samples was measured by the RAD7 RAD H2O system (Durrige Co.). Part of the entire measuring system is an air pump that continuously exhales radon within 5 minutes through the closed loop aeration process of the water sample. In this process, almost 94% of radon was extracted from the sample. After that, the pump is automatically stopped, and then the system is idle for 5 minutes, in order to restore the balance between water, air and radon products in the detector. The lower detection limit of the device is 0.37 Bq/l. So, as the true value of radon concentration in water samples, its corrected value is used according to the formula (Todorovic et al., 2012):

$$C_{Rn} = C_o \bullet \delta. \quad (1)$$

where: $\delta = e^{\lambda t}$, where for radon decay $\lambda = 0.00756\ h^{-1}$, C_o defines value of radon concentration in water sampling measured in laboratory and t defines the time elapsed from sampling to laboratory analysis. The time elapsed from sampling to analysis was 4 days.

As the population uses water in the household, the contribution of radon from the water to the closed space in the room was calculated, according to the pattern (Zelewski et al., 2001):

$$C_{aRn} = C_{wRn} \times W \times \frac{e}{(V \times \lambda_c)}. \quad (2)$$

where: C_{aRn} is the contribution of radon from water to the total radon in the air, C_{wRn} is the activity concentration of radon in water, W is the average intake of water (0.01 m^3/h per person), e is the coefficient of exhalation of radon from water into air (0.5), V is the volume of the room (30 m^3), and λ_c is the coefficient of the change of air in the room (0.7 h^{-1}) (UNSCEAR; 1993; Xinwei, 2006).

The carcinogenic effects of radon, in the long term, is reflected in the determination of the total effective dose of internal irradiation with radon dissolved in water, which consists of two components: the first is defined by the effective dose of radon inhalation, while the second is defined by the effective dose of radon ingestion. According to the 2000 UNSCEAR Report, these quantities can be determined as follows:

-*The annual effective doses for inhalation* waterborne radon were calculated by using the parameter established in the UNSCEAR 2000 as (Somlai et al., 2007; El-Araby et al., 2019; Deeba et al., 2021):

$$E_{inh}\left(\frac{\mu Sv}{y}\right) = C_{wRn} \times C_{aw} \times DCF \times F \times t. \quad (3)$$

where: E_{inh} is the effective dose for inhalation, C_{WRn} is the radon concentration in water (Bq/l or kBq/m^3), C_{aW} is the radon in air to the radon in water ratio (10^{-4}), F is the equilibrium factor between radon and its progenies (0.4), t is the average indoor occupancy time per individual (7000 h/y) and DCF is the dose conversion factor for radon exposure (9 nSv (Bq h $m^{-3})^{-1}$).

According to data presented in UNSCEAR (2000), the transfer factor C_{aw} has an order magnitude of 10^{-4}. As part of these investigations, the exact value of the ratio of radon concentration in water and its contribution in air was determined, so the calculated value presented in Table 1. is used in equation 2.

-*The annual effective doses for ingestion* radon were calculated using the following formula (UNSCEAR, 2000):

$$E_{ing}\left(\frac{\mu Sv}{y}\right) = C_{wRn} \times C_w \times EDC. \quad (4)$$

where: E_{ing} is the effective dose for ingestion, C_{wRn} is the radon concentration in water (Bq/l or kBq/m^3), C_w is the the weighted estimate of water consumption (60 l/y) and EDC is the effective dose coefficient for ingestion (3.5 nSv/Bq), respectively.

RESULTS OF RESEARCH

The summarized results of the research in this work are presented in 2. In addition to temperature and radon concentration in samples from 12 selected localities, the contribution of radon from water to the total radon in the air in closed rooms is also presents in Table 2. In the last column, there is C_{aw} - the calculated value of the transfer factor within these studies and which is used in equation 2 for determination the annual effective dose from radon inhalation.

Table 2. Summarized results of research.

No	T (°C)	C_{wRn} (kBq/m^3)	C_{aRn} (Bq/m^3)	$C_{aw} = C_{aRn}/C_{wRn}$ (10^{-4})
1	8	8.7±1.4	2.6±0.4	2.9
2	12	6.6±1.6	2.0±0.5	3.0
3	12	13.8±3.6	4.1±1.1	2.9
4	14	19.1±1.4	5.7±0.4	2.9
5	13	21.5±2.5	6.5±0.7	3.0
6	14	11.8±2.9	3.5±0.9	2.9
7	12	6.6±1.2	2.0±0.3	3.0
8	12	6.8±1.4	2.1±0.4	3.0
9	13	2.8±1.2	0.8±0.3	2.8
10	11	6.4±1.7	1.9±0.5	2.9
11	22	76.0 ±4.0	22.8±1.2	3.0
12	13	4.3±0.8	1.3±0.2	3.0
Min	8	2.8±1.2	0.8±0.3	2.8
AV	13	15.3±2.0	4.5±0.6	2.9
Max	22	76.0±4.0	22.8±1.2	3.0

In order to correctly determine the contribution of radon from water to the total radon in the air in a closed room, instead of Bq/l, the radon concentration in water is expressed in kBq/m^3, whereby a radon concentration in water of 1mBq/l corresponds to a radon concentration of 1Bq/ m^3 in air.

According to the temperature range from 8°C to 22°C, these underground waters can be classified as cold waters. Intervals of measured values of radon concentration in water ranged from 2.8±1.2 kBq/m^3 to 76.0±4.0 kBq/m^3, with an average value of 15.3±2.0 kBq/m^3. Radon concentration values in the sampled waters are relatively low. All values are below the recommended WHO (2004) value. This could be expected if one takes into account the geological structure of the place where the springs are located (Table 2). According to the presented data, it can be said that the waters from these areas are radiologically correct and can be used for drinking, since their values are far below the recommended value of 100 kBq/m^3 (WHO, 2004). In seven samples radon concentration in the water was below 11 kBq/m3 on seven sites, which is the upper limit of the radon concentration in drinking water (Fig 3) according to the recommendation of the US-EPA (1999).

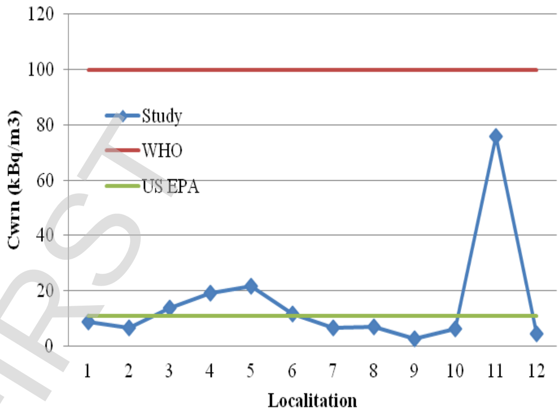

Figure 3. Range of radon concentration in water related to recommended values.

Considering the geological structure of the places where the sources of the sampled water are located, the presented results are expected (Table 1). The terrain itself does not contain minerals and rocks containing radionuclides, primarily uranium and radium, so radon concentrations are low. There was only location 11 with a detected higher radon concentration (76.0±4.0 kBq/m^3), but the geology of the place itself does not point to that. This pronounced concentration may be a consequence of the movement of groundwater over rocks rich in radon. Groundwater dissolves rocks and carries dissolved radon with it on its further way. This could be the reason why there is a higher concentration of radon at location 11. Concentration of radon in water measured in this research are comparable with the data of other researchers (Khandaker et al., 2020; Mehnati et al., 2022; Kumar et al., 2022; Vučković et al., 2022).

As radon is a radionuclide that easily leaves water, water with an elevated radon level can cause radon diffusion into the interior of the room and lead to an increase in the total radon level. This leads to the conclusion that the population from these localities is exposed both ingestion and inhalation of radon from water. The interval of certain values of radon concentration that is released from the water, and thus increases the total concentration of radon in closed rooms, is from 0.8±0.3 Bq/m^3 do 22.8±1.2 Bq/m^3, with mean value of 4.5±0.6 Bq/m^3.

The value interval of the transfer factor C_{aw} is from 2.8x10^{-4} to 3x10^{-4} and corresponds to the recommended range of transfer factor values from 10^{-5} to 3x10^{-4} (paragraph 93 in UNSCEAR 2000).

When consuming radon-rich water, the first cells to be exposed to alpha particle irradiation emitted during the decay of radon and its short-lived progeny are gastric tissue cells (WHO, 2004). During inhalation of radon and its decay products, lung tissue is exposed to direct radiation. Based on this, the total annual exposure of the population to radon from water can be determined through: the annual effective dose of inhalation (E_{inh}) and the annual effective dose of radon ingestion (E_{ing}). Table 3. shows the values of the annual effective doses of inhalation and ingestion of radon present in water.

Table 3. Estimation of radiation doses of population to radon from water on the selected sites.

No	E_{inhal} (μSv/y)	E_{ingest} (μSv/y)	Doses to organs (μSv/y	
			lungs	stomach
1	63.6±12.4	1.8±0.3	7.6	0.2
2	49.9±12.1	1.4±0.4	5.9	0.1
3	100.8±26.3	2.9±0.8	12	0.3
4	139.5±10.2	4.0±0.3	16.7	0.5
5	162.5±18.9	4.5±0.5	19.5	0.5
6	86.2±21.2	2.5±0.6	10.3	0.3
7	49.9±9.1	1.4±0.2	5.9	0.2
8	51.4±10.6	1.4±0.3	6.2	0.2
9	19.8±8.5	0.6±0.2	2.4	0.07
10	46.7±12.4	1.3±0.4	5.6	0.1
11	574.5±30.2	16.0±0.8	68.9	1.9
12	32.5±6.0	0.9±0.2	3.9	0.1
Min	19.8±8.5	0.6±0.2	2.4	0.07
AV	114.8±14.8	3.2±0.3	13.8	0.4
Max	574.5±30.2	16.0±0.8	68.9	1.9

The interval in which the annual effective inhalation dose ranges is from 19.8±8.5 μSv/y to 574.5±30.2 μSv/y. The mean value of the annual effective dose of radon inhalation is 114.8±14.8 μSv/y, and it corresponds to the recommended value of 100 μSv/y (WHO, 2004). The extreme value of 574.5±30.2 μSv/y measured in sample 11 is a consequence of the increased presence of radon in the water. In all other locations (1, 2, 6, 7, 8, 9, 10 and 12) the annual effective inhalation dose is below the recommended value, while the inhalation dose values in locations 3, 4 and 5 apears to be close to the recommended value of 100 μSv/y. Range of annual effective doses of ingestion is from 0.6±0.2 μSv/y to 16.0±0.8 μSv/y, while a mean value is 3.2±0.3 μSv/y. Mean value of the total annual effective dose of exposure is 118.0±15.3 μSv/y. In the special case, that no other organ is exposed the dose contribution from this source to the lungs and stomach is calculated by multiplying the inhalation and ingestion dose with tissue weighting factor (w_t) of 0.12 for lungs and stomach, respectively (UNSCEAR 2000). Two last colons in Table 3. represent the radiation dose contribution of radon in drinking water to lung and stomach. Mean values of these doses are 13.8 μSv/y and 0.4 μSv/y, respectively. Based on all the presented values of the selected parameters, it can be concluded that the selected localities can use for consumption and for other domestic usage from the point of view of radiation protection.

CONCLUSION

Research presented in this paper includes 12 carefully selected localities where the population is supplied with drinking water from natural captured sources. The research included only those households that are very similar in terms of their habits and the spatial organization of the houses they live in. Interval of radon concentration present in water is from 2.8±1.2 kBq/m^3 to 76.0±4.0 kBq/m^3, with a mean value of 15.3±2.0 kBq/m^3. As the measured values are below the recommended level of 100 kBq/m^3 (WHO, 2004), the waters from these localities are safe for wider household use. Contribution of radon from water to radon in the air in closed rooms is llow, but it should be taken into account because natural springs are the only way to supply drinking water in this area. Mean value of the calculated transfer factor is 2.9x10^{-4} and corresponds to the generally accepted parameter from the literature (UNSCEAR 2000). According to the average value of the annual effective dose of inhalation and ingestion radon from water, it can also be said that waters in these areas are safe for use both for drinking and for other numerous household needs.

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