

Radon activity assessment of thermal water in Spas of Kosovo by using different methods

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► Short report

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INTRODUCTION

Radon (^{222}Rn), a progeny of uranium (^{238}U), is a colorless, odorless, and ubiquitously present but noble gas that is radioactive and poses grave health hazards not only to uranium miners but also to people living in households and flats, as well as at a workplaces in the industry. Its most stable isotope, ^{222}Rn , has a half-life of 3.8 days (¹⁻²). As radium decays, radon is formed and is released into small air or water-containing pores between soil and rock particles. It usually migrates freely through faults and fragmented soils and may accumulate in caves or water (³).

Radon is one of the densest substances that remains a gas under normal conditions and is considered a health hazard due to its radioactivity (⁴).

Radon is emanated from solid grains and propagates through the soil pores, filled with air and water and then it is transported by diffusion and advection through this space and is exhaled into the atmosphere. Exhalation rates are highly affected by the permeability of the soil and by atmospheric parameters, such as rainfall, real humidity, temperature, and barometric pressure (⁵⁻⁶).

High ^{222}Rn concentrations occur in groundwater

ABSTRACT

Background: Exposure to radon and radon decay products at home and at workplaces constitutes one of the greatest and perhaps the greatest risk from ionizing radiation. In this study, radon activity concentrations were measured in thermal water, spring water, in the water of baths/pools, and indoor air of five spas in Kosovo. **Materials and Methods:** Different comparison active methods were used and also the long-term measurements were performed. **Results:** The radon activity concentrations were found to be 23 Bq l^{-1} to 314 Bq l^{-1} in water coming from a thermal source, 13 Bq l^{-1} to 270 Bq l^{-1} in water of baths, 25 Bq l^{-1} to 108 Bq l^{-1} , in water of pools and 16 Bq l^{-1} to 67 Bq l^{-1} in taps water. The indoor radon concentrations and soil gas measurements near the thermal spas has range from 24 Bq m^{-3} to 506 Bq m^{-3} respectively from 4978 Bq m^{-3} to 67249 Bq m^{-3} . **Conclusions:** The indoor air radon concentrations in workrooms, baths, and pools only in one case exceeded 500 Bq m^{-3} . The reason of high indoor radon concentration could have been the location of this room, located near the baths and pool of spa. The maximum radon activity concentration in taps water is the only that did not exceed the limit from 100 Bq l^{-1} .

in many areas where wells are used for domestic water supply and thermal spas (⁷⁻⁸).

Radon and decay product nuclei are present in indoor environments of spa facilities and have been identified as an agent of additional radiation for bathers and working personnel (⁷⁻⁸).

Radon can present a hazard in a wide range of workplaces other than mines. While this includes below ground workplaces such as subways, tunnels, stores, show caves, closed-out mines open to visitors, and radon spas, the majority of such workplaces will be above ground (⁹).

The World Health Organization (WHO) set the guidance level to 100 Bq l^{-1} for water radon concentration in the third edition of the WHO drinking water guidelines (¹⁰).

Several surveys for presence of radon in spas were performed in Europe to evaluate the indoor radon levels and exposure to radon. Radon measurements of thermal waters from Łądek Zdrój in Poland presents values from 122 Bq l^{-1} to 1284 Bq l^{-1} . Radon concentrations in thermal waters of spas in Lesvos Island of Greece were found among range 10 and 304 Bq l^{-1} . Radon measurements of air and thermal water in nine Croatian spas was conducted and radon concentrations in the air and water of the

swimming pools was obtained at the range of 10.9–109 Bq·m⁻³ and 0.73–18.6 Bq l⁻¹, respectively. Radon measurement in five Slovenian spas was conducted. Indoor radon was obtained in the range of 15–279 Bq·m⁻³⁽¹¹⁾.

The thermal spas in Kosovo, has been used traditionally for purpose of medical rehabilitation. Many local population and foreign visitors had a visit and have received treatment in these spas. In the Klokot spa center, there are unused resources of mineral water at 37 °C and the healing mud known as Peloida of Klokot spa. The temperature of the mineral water in spa of Peja ranged from 11 °C to 47 °C.

In addition to the indoor and outdoor pools used by patients, in these spa centers, the service of physiotherapy with contemporary treatment is as well available.

This study has a aim to give a picture of more complete situation about the radon doses that are received by the staff and visitors of Kosovo spas and health risks due to exposure of radon. These results will contribute in establishment of a national basis and standards and will help the institutions to establish a national radon strategy and the action plan. The study itself will contribute to know the radon activity concentrations in indoor air, in water, and in soil gas of Kosovo spas and to provide recommendations for monitoring of the dose received by working staff and visitors of spas.

MATERIALS AND METHODS

Site description and samples collection

There are 5 thermal spas in Kosovo, located in different areas of the country, which are providing health services for patients and visitors. The locations of all five spas in Kosovo are presented in below given figure 1, by colored points.

The spas are known for their health rehabilitation and for curing rheumatism diseases, neurological diseases, traumatically, cardiovascular, dermatological, gynecological, and similar diseases. It is known throughout the public, not only in Kosovo for quality healing affects, but also having in mind and being grateful primarily for multiple infinite resources as those of thermo-mineral water alike pellucid (mud) also with effect of great healing.

The water measurement of radon concentrations was carried out with the method of alpha scintillation cells and Alpha GUARD system.

Instantaneous radon concentration was measured by alpha scintillation cells (0.7 dm³) manufactured at the Jožef Stefan Institute and calibrated by a standard ²²⁶RaCl₂ solution (National Institute of Standards and Technology (NIST), Standard Reference Material 4953D), according to the Rushing procedure. Cell efficiency is about 1.4 x 10⁻³ s⁻¹ Bq⁻¹ m³ which, at

30–60 min counting time, gives a lower limit of detection of 10–20 Bq m⁻³ at 1–2 min⁻¹ background⁽¹²⁾.

With the scintillation cells method (PRM - 145 device, produced by AMES, Ljubljana, Slovenia) the water samples were taken using 250 and 333 ml glass bottles. Allowing the water flow for some time from the spring/well source after the bottle was filled with a slight "bulge" of water standing above the rim of the bottle. In case there were seen air bubbles in the bottle, the same bottle was refilled again. The samples from bathtubs/public pools water, was not taken on the water surface but in a depth of water, which produce the most accurate results. For each sampling, the location, time and date of sampling was evidenced.

Air from the water was sampled directly into a cell, then a cell was transported to the laboratory, and gross alpha radiation counted after 3 hours, when the equilibrium between radon and its decay products was reached⁽¹²⁾.

A radon concentration of the water sampling was measured also by using the professional radon monitor AlphaGUARD PQ2000 PRO, which is a portable radon monitor with high storage capacity. It is an ionization chamber (0.62 l), designed for measuring radon in air, soil, and water. The AquaKIT, is the additional equipment of AlphaGUARD, which was used for the water measurements⁽¹³⁾.

AlphaGUARD has an ionization chamber that is also a part of the gas cycle. Radon was expelled from water samples (placed in emanation vessel) by using the pump in a close gas cycle. The safety vessel was connected with the emanation vessel. All drops are deposit in it, if they had got into the gas cycle during the degassing process. This way the pressure of the water vapor was minimized for the radon monitor. After that, the water was injected into the emanation vessel, and the AlphaGUARD and AlphaPUMP were switched on. The flow rate of the pump was 0.3 l min⁻¹⁽¹³⁾.

With the AlphaGUARD method after, 10 min., the pump was switched off and the AlphaGUARD remained switched on for another 20 min, so the radon measurement has continued. The AlphaGUARD monitor worked in a 'flow' mode for 1 minute. Prior to every water sample measurement, for a few minutes, the background of the empty set-up was measured. Calibration of the measuring system has been carried out by Saphymo (Genitron Instruments, Germany), with guaranteed stability and accuracy for 5 years⁽¹³⁾.

Calculation of radon concentration in the measured water sample with Alpha GUARD System was based on equation (1):

$$C_{\text{water}} = \frac{C_{\text{air}} \times \left(\frac{V_{\text{system}} - V_{\text{sample}}}{V_{\text{sample}}} + k \right) - c_0}{1000} \quad (1)$$

- C_{Water} - Rn concentration in water sample [Bq l⁻¹];
 C_{Air} - Rn concentration in the measuring set-up (indicated by Alpha GUARD) [Bq m⁻³];
 C_0 - Rn concentration in the measuring set-up before sampling [Bq m⁻³];
 V_{System} - interior volume of the measurement set-up [ml];
 V_{Sample} - volume of the water sample [ml];
 k - radon diffusion coefficient;

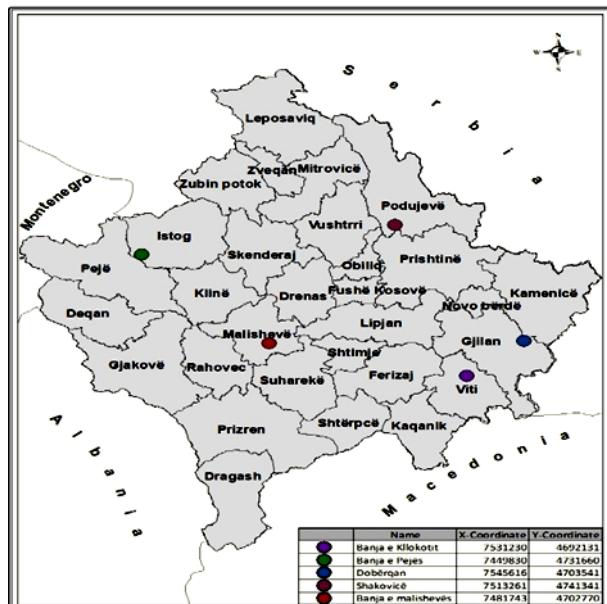


Figure 1. Kosovo map location of spas.

The measurement of indoor radon concentration was performed by exposing solid-state nuclear track detectors CR-39 (Radosys, Veszprem, Hungary). The CR-39 detectors were provided and evaluated by the Institute of Radiochemistry and Radioecology at the University of Pannonia, Hungary⁽¹⁴⁾.

The exposure time of CR-39 detectors in most of the locations was around three months, including at least one month in a winter time. At all points, detectors were placed on the ground floor, preferably in the frequented room, where the workers or visitors of spa, spent most of their time indoors. They were fixed at 1–1.5 m above the floor and more than 0.5 m away from any wall as well from any other object⁽¹⁴⁾.

These measurements were performed for indoor radon concentrations in staff rooms, baths, and pools. In some points near the spas, also the soil gas measurements using track detectors was conducted.

Statistical analyses

Basic statistical analyses were performed using Excel version 2013. These statistics include maximum and minimum values, mean, correlation, standard deviation. Maximum value of water radon

concentrations and indoor radon concentrations was 314 Bq l⁻¹, respectively 506 Bq m⁻³ and minimum 22 Bq l⁻¹ respectively 24 Bq m⁻³. An indoor radon concentration was in good correlation with water radon concentrations. The mean value for radon activity concentrations in thermal water was 286 Bq l⁻¹ with standard deviation \pm 25 Bq l⁻¹, in water of baths/pools 272 Bq l⁻¹ with standard deviation \pm 20 Bq l⁻¹, in pool's water 105 Bq l⁻¹ with standard deviation \pm 5 Bq l⁻¹ and in tap's water from 51 Bq l⁻¹ with standard deviation \pm 14 Bq l⁻¹.

RESULTS

A significant number of 40 water samples were collected, from a thermal source, in individual bathtubs, public pools, and in taps water.

Indoor radon concentrations was measured in 27 locations in doctor's rooms, patient's rooms, pools, and baths ranged from 24 Bq m⁻³ to 506 Bq m⁻³.

The radon concentration values in water from a thermal source, in individual bathtubs, public pools, and in taps water of spas ranged from 23 Bq l⁻¹ to 314 Bq l⁻¹ in thermal water, 13 Bq l⁻¹ to 270 Bq l⁻¹ in the water of baths, 25 Bq l⁻¹ to 108 Bq l⁻¹ in the water of the pool and 16 Bq l⁻¹ to 67 Bq l⁻¹ in taps water, respectively.

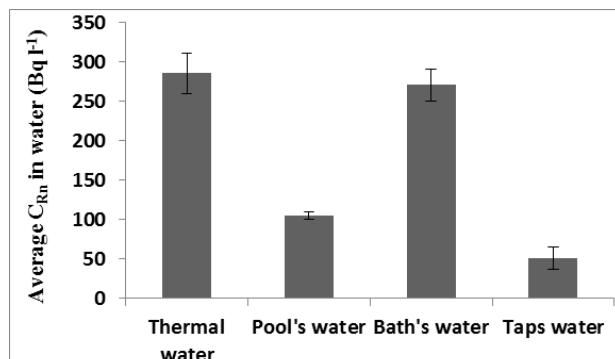


Figure 2. Average radon concentrations in water from different places.

In above figure 2, the maximum average of radon concentrations was found in thermal water 286 ± 25 Bq l⁻¹, comparing with water of baths/pools 272 ± 20 Bq l⁻¹ and pool's water 105 ± 5 Bq l⁻¹. The minimum average of radon concentration from figure 2 was found in tap water from 51 ± 14 Bq l⁻¹.

The highest value of radon concentration, 314 Bq l⁻¹ was found on hot spring water in the spa of Shakovica. The highest value of radon concentrations on individual bathtubs/public pools and in the pools with water was found in the spa of Kllokot, 270 Bq l⁻¹, respectively 108 Bq l⁻¹. And the highest value of the radon concentration in the drinking water was evidenced in the spa of Dobërçan, 67 Bq l⁻¹.

The above figure 3 shows the correlation of radon activity concentrations in indoor air with radon

concentrations in water. At the locations where radon was high in the water, also the indoor radon was increased.

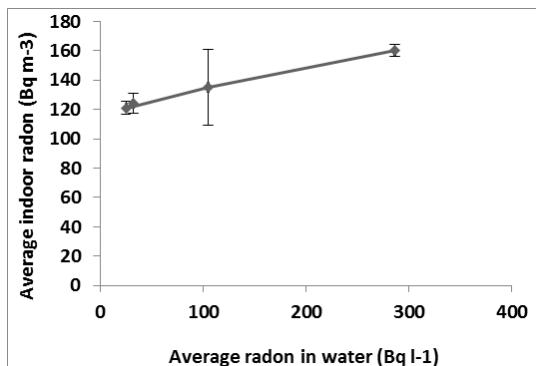


Figure 3. Correlation between indoor radon and radon in water.

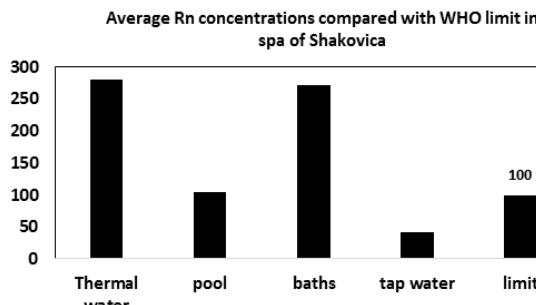


Figure 4. Comparison of average values with the WHO limit.

DISCUSSION

In most of the rooms, indoor radon concentrations was below the approved limit in the legislation of Kosovo and European Basic Safety Standards (EU BSS) from 300 Bq m^{-3} (15), except in one case where the radon activity concentrations exceeded 500 Bq m^{-3} . This high indoor radon concentration can be explained by the fact that this room is located near the pool and the baths of the spa. But this value is less than in other countries as in V4 countries, where the highest indoor radon concentrations was more than $12\,000 \text{ Bq m}^{-3}$ and higher than in Hungarian thermal baths where radon concentrations was below the recommended level from EU BSS of 300 Bq m^{-3} (15).

The average radon concentrations in thermal water $286 \pm 25 \text{ Bq l}^{-1}$, the water of baths/pools $272 \pm 20 \text{ Bq l}^{-1}$, and in pool's water $105 \pm 5 \text{ Bq l}^{-1}$ are higher than the limit recommended by WHO (10) of 100 Bq l^{-1} . Only the radon concentration in tap's water from $51 \pm 14 \text{ Bq l}^{-1}$ is below the limit recommended by WHO of 100 Bq l^{-1} , fig.4.

The maximum level of radon concentrations in thermal water 314 Bq l^{-1} in comparison with other countries is higher than in Greece from 304 Bq l^{-1} (16), Croatian from 18.6 Bq l^{-1} and in spas of Turkey from 5.89 Bq l^{-1} (17-18) and lower than in Poland (19) from

1284 Bq l^{-1} and V4 countries spas from 384 Bq l^{-1} (11).

Also, the highest radon concentration in baths and public pool water of the spa in Kllokot from 270 Bq l^{-1} respectively 108 Bq l^{-1} , was lower than in thermal spas of V4 countries from 383 Bq l^{-1} , respectively 373 Bq l^{-1} and in Portuguese thermal spas and higher than the limit recommended by WHO (10) and United Nations Scientific Committee on the Effects of Atomic Radiation (20) from 100 Bq l^{-1} .

The level of radon concentrations in indoor air and water as well may be caused by different factors. These factors can be geological structures of the zone, meteorological parameters, the number of radioactive elements content in soil, respectively in underground water, etc.

Because this is the first research in thermal spas of Kosovo, the extended studies are recommended and monitoring of dose received by staff of spas must be regularly checked according to EU BSS recommendations (15).

CONCLUSION

Only the maximum radon concentration that was measured in drinking water was 67 Bq l^{-1} , below the limit recommended by WHO and UNSCEAR 1993 of 100 Bq l^{-1} . The maximum value of indoor radon concentration only in one room was found to be 500 Bq m^{-3} , which is higher than the limit recommended by EU BSS of 300 Bq m^{-3} .

High indoor and water radon concentrations can be caused by high ^{222}Rn concentrations that occur in groundwater used for thermal spas and geological structures of the zones, and not considered to be affected by soil gas radon concentrations.

This study will contribute to know the level of exposure to radon from staff, visitors, and patients in thermal spas in Kosovo due to exposure in indoor air and from inhalation and ingestion of radon from water.

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Conflicts of interest: Declared none.

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