RADON LEVELS IN PORTUGUESE THERMAL SPAS*

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Abstract. *Radon concentration measurements were performed in indoor air and in natural mineral waters in seventeen Portuguese thermal spas used for therapy, drinking and irrigation purposes. The gamma doses rates were also assessed in different workplaces of the considered thermal spas. The radon concentration was measured in water samples taken from different sampling points: boreholes, springs, inhalator chambers (ORL's) and swimming pools, and in the indoor air of different treatment rooms: ORL's, swimming pools, vapours areas and Vichy shower. Radon concentration in water ranged from 26 to 6949 Bq/L and in the indoor air ranged from 73 Bq/m3 to 3479 Bq/m3. The indoor gamma dose rates ranged from 0,148 µSv/h to 0,644 µSv/h and the annual dose rate was estimated ranging from 0,30 to 1,29 mSv/y, for 2000 working hours per year, which is far below the effective dose limit for workers (20 mSv/y). Nevertheless, the great contribution for the annual effective dose will result from radon inhalation which is not included in this estimation. The remedial action level for drinking water of 1000 Bq/L (2001/928/ EURATOM) was exceeded in 23% of the selected thermal spas and about 80% of the total measurements of the indoor radon concentration exceeded the previous reference level of 200 Bq/m3 for new buildings and about 63% exceeded the new reference level of 300 Bq/m3 as established in the Directive 2013/59/EURATOM for indoor radon concentration in workplaces. Therefore, as the recommended limits for radon concentration in water and in indoor air were exceeded, appropriate actions should be taken in order to reduce the hazard to health from radon indoors and the potential resulting occupational exposure.*

Key words: *radon, thermal spa, dose rate, occupational exposure*

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1. INTRODUCTION

Radon gas is recognized as the most significant natural source of human exposure and the leading cause of lung cancer incidence, with the exception of tobacco [1, 2, 3, 4, 5, 6]. Thermal spas have been identified as one of the professional activities with potentially higher exposure to radon mostly due to the inhalation of radon released from thermal waters [7].

Radon concentration in groundwater varies considerably (1 - 10 000 Bq/L) depending mostly on the uranium concentration in the surrounding rock and on the water circulation [8]. Radon's solubility in water increases with decreasing temperatures and therefore much attention has been given to thermal water sources due to the potential risk to the public health. Moreover, radon concentration levels in groundwater are generally higher than in surface water [3].

Human exposure to ^{222}Rn and its decay products in water supplies can occur in two ways: by ingestion (drinking water) or by inhalation due to the release of radon into the air when using thermal waters for health purposes [9]. The risk of radon exposure is mostly associated with high radon concentrations in confined environments, and the subsequent inhalation, increasing the risk of damaging the organ cells where radon short-life products are deposited.

Several studies have been conducted worldwide to measure the concentration of radon in the water and in the air of thermal spas in order to estimate the exposure doses both for workers and for users [8, 10, 11, 12, 13, 14, 15]. Recently, a study carried out in thermal spas from Ischia Island showed that the average annual effective dose to workers due to radon exposure (3,52 mSv/y; range 0,01-7,03 mSv/y) was higher than the dose limit imposed by Italian legislation (3 mSv) [16].

The health effects of exposure to radon in indoor environments (indoor air), and in particular, in thermal spas, depend mainly on the concentration of inhaled radon, the ventilation rate of the place, freq uency and duration of the exposure [7, 17]. In Portugal, the monitoring of radon concentration is compulsory only in buildings constructed in granite areas, particularly in the districts of Braga, Vila Real, Porto, Guarda, Viseu and Castelo Branco, due to the geological characteristics of these regions. This obligation came into force in 2006 within the new regulations for indoor air quality imposed by the Portuguese legislation [18].

The purpose of this work was to measure the radon concentrations in indoor air and in natural mineral water as well as to evaluate the gamma doses rates in different workplaces of Portuguese thermal spas.

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2. METHODS

Seventeen thermal spas participated in this study, representing about 46% of all thermal spas existing in Portugal (Fig. 1). For each studied thermal spa, the radon concentrations were measured in the natural mineral water from different sampling points: boreholes (BH), springs (SG), inhalator chambers (ORL´s) and swimming pools (SP) and in the indoor air of different rooms: ORL's, swimming pools (SP), vapours areas (VA) and *Vichy* shower (VS). The measurements were carried out between November 2013 and September 2014. The assessment of radon concentration included measurements in two different periods (summer and winter) in the indoor air of the selected thermal spas. Regarding the assessment of radon concentration in water a single measurement was carried out directly in the water from the borehole and in the water used in the treatment rooms. Measurements of gamma radiation doses were also assessed in some of the previous locations.

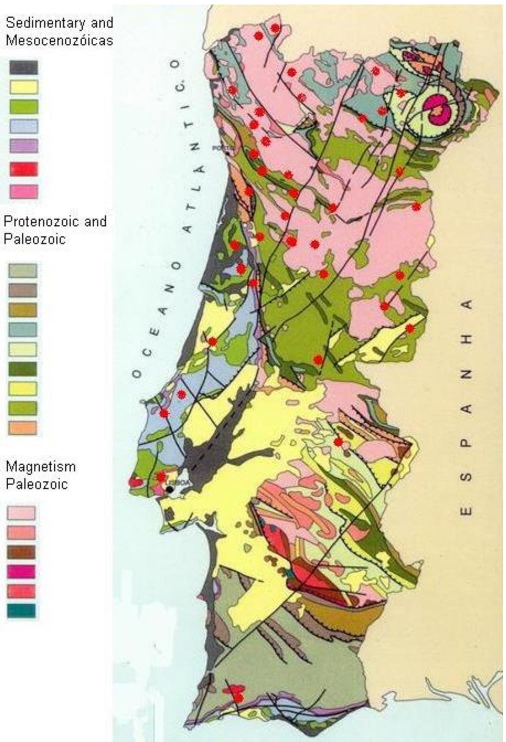

Figure 1. Location of the Portuguese thermal spas

Radon concentration in water was measured by liquid scintillation. The samples were sent to the Natural Radioactivity Laboratory of the Department of Earth Sciences, University of Coimbra, Portugal, for measurement by a liquid scintillation counter. The radionuclide activity was measured by liquid scintillation counting techniques (LSC) using an ultra-low-level spectrometer (Quantulus 1220). For radon measurements the samples were collected in glass containers of 20 mL previously filled with 10 mL of a water immiscible scintillation cocktail, with sealed opening and security mechanisms to avoid gas-leakage during transport. With this protocol radon was measured through the double-phase method [19].

Radium and uranium isotopes were measured in water samples, previously filtered, acidified with HNO_3 to pH less than 2 and, only in the former isotope, pre-concentrated by evaporation. The ^{226}Ra activity was measured through the indirect method based on radon accumulation in a lipophylic scintillation cocktail during a minimum of 21 days (secular equilibrium). Before LSC counting a selective extraction of uranium was performed using an "extractive cocktail" (bis-2-etilhexyl-ortophosphoric acid) and a non-water soluble scintillation cocktail. After phase separation the cocktail, collected in polyethylene vials, is measured from the peaks of ^{238}U and ^{234}U. This method has been tested by comparison with other methods (like ICP-MS) and the results shows good agreement [20].

The uncertainties depend on the activities but, for the range of values measured, were generally lower than 15%. For a more detailed description about the LSC techniques, see [21]. Efficiency was evaluated by measuring several standard solutions and participation in intercomparison exercises.

Radon concentration measurements in air were performed using CR-39 detectors enclosed in small cylindrical (5-cm height, 3-cm diameter) diffusion chambers for periods between 25 and 45 days. This detector is a small piece of plastic that is sensitive to tracks of highly ionizing particles such as alpha particles. The CR-39 detectors were placed in each room at approximately 2 meters from the floor (breathing zone) [22]. After a period of exposure between 25 and 45 days, the detectors were retrieved and sent to the Natural Radioactivity Laboratory of the Department of Earth Sciences, University of Coimbra, Portugal.

Indoor gamma dose rates were performed by a Geiger counter (Gamma Scout) which is a calibrated measurement instrument for alpha, beta and gamma rays. This device was used for the measurement of gamma doses rates which was hourly acquired and stored for a time period between 25 and 45 days. Since this system acquires the dose continuously the variation of dose during different periods of time is averaged out. Therefore, long-term integrated measurements of gamma dose may be taken.

3. RESULTS AND DISCUSSION

3.1. Radon concentration in natural mineral water

The results of radon concentration in natural mineral water samples taken from each one of the studied thermal spas (TS) are presented in Fig. 2 and Fig. 3. Radon concentrations in natural mineral water from ORL's range between 26 Bq/L and 5325 Bq/L (Fig. 1). About 33% of the water samples taken in ORL's from the considered thermal spas exceeded the action level for radon concentration recommended by the European Commission 2001/928/EURATOM, establishing for concentrations in excess of 1000 Bq/L, remedial action is deemed to be justified on radiological protection grounds. In what concerns to water samples taken from the ORL's of TS7, the values of radon concentration are very different: 2624 Bq/L

(ORL 7.1) and 1461 Bq/L (ORL 7.2). This can be explained by the different type of treatment (water utilization) in each one of these ORL's. In addition, another important variable is that TS7 has an operating ventilation system which may function in two ways: first to exchange air with the outdoor and secondly, to create over-pressurised air in order to avoid the radon intrusion into the ORL's. The greater the pressure differential the higher the radon level which suggests that ORL 7.1 presents a higher pressure differential than ORL 7.2.

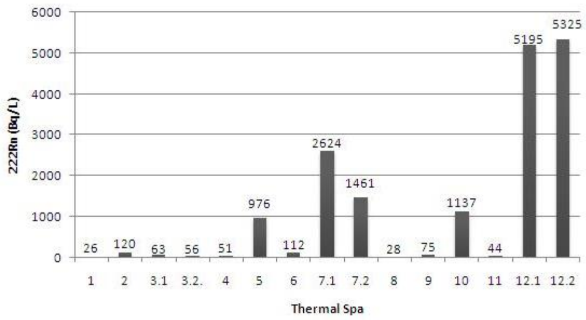

Figure 2. Radon concentration in water samples taken from the ORL's chambers (Bq/L)

In what concerns to water samples taken from the boreholes (BH) (Fig. 3), the concentration of radon is higher than in the water from the ORL's, ranging between 41 Bq/L (TS1) and 6949 Bq/L (TS17, BH 17.1). However, only in 25% of the water samples, taken from boreholes of the 17 studied thermal spas, the concentrations were excess of 1000 Bq/L. The high concentrations of radon found in TS11 and TS17 may be explained by local the geological settings, a bedrock composed by metasedimentary rocks with high uranium and thorium occurrences [13, 14].

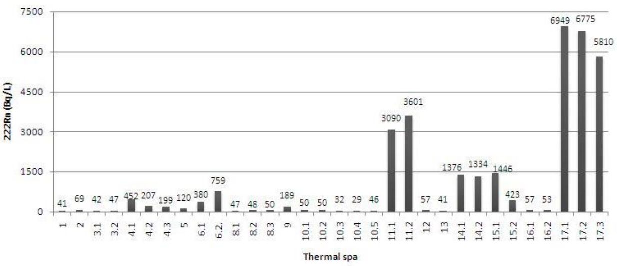

Figure 3. Radon concentration in water samples taken from the boreholes (Bq/L)

The radon concentration was measured in the water from the buvettes of 2 thermal spas which may be used for ingestion depending on the medical prescriptions. The obtained results, 59 Bq/L for TS1 and 973 Bq/L for TS2, only the TS1 did not exceed the reference level recommended by the European Union (100 Bq/L). Nevertheless, this difference is mostly due to the geological settings and consequently different characteristics of the thermal water from each one of these thermal spa.

The concentration of radon was also analysed in water samples taken from the springs of 3 thermal spas. Only one result exceeded the reference level recommended by the European Union (100 Bq/L): 23 Bq/L (TS1), 86 Bq/L (TS2) and 478 Bq/L (TS3).

3.2. Radon concentration in the indoor air

The results of radon concentration measured in the indoor air for each one of the studied thermal spas (TS) are presented in Table 1.

Table 1 Radon concentration in indoor air

TS	Sampling location	^{222}Rn (Bq/m^3)
1	Technical area	422
	Medical Office	577
	Thermal pool	355
	ORL	707
	Hall Spa	841
2	Technical area	481
	Thermal pool	618
	Access corridor to the thermal pool	1079
	Ludic pool	641
3	ORL	375
	Steam hall	398
	Bathtubs	172
	Thermal pool	370
	Sludge area	467
4	Rehabilitation pool	813
	Thermal pool	862
	Technical area	1692
5	Thermal pool	517
	Treatment area	692
	Vichy shower	724
	ORL	329
	Access corridor to the thermal pool	566
6	Thermal pool	449
	ORL	3479
	Vichy shower	674
	Thermal pool	784
7	ORL	502
	Thermal pool	274
	ORL	401
	Vapours	453
	Vichy shower	437
8	ORL	169
	Vichy shower	406
	Thermal pool	121
9	ORL	312
	Access corridor to the thermal pool	116
	Vichy shower	112
	Thermal pool	73
10	Jet shower	1130
	ORL	2298
	Vichy shower	1971
	Technical area	1145
	Thermal pool	1494
11	ORL	146
	Thermal pool	203
	Vichy shower	93
12	ORL	169
	Vichy shower	376

The highest values of radon concentrations in indoor air were registered in the ORL's of TS6 and TS10, followed by the *Vichy* shower of TS10 and technical area of TS4, which are above of the reference level of 300 Bq/m^3, as published in the Directive 2013/59/EURATOM, laying down the basic safety standards for protection against the dangers arising from exposure to ionizing radiation. These thermal spas have only natural ventilation (no mechanical) and are inserted in a radon prone area (with a geological setting mainly comprised by granites) in addition to the presence of a dense network of tectonic fractures intersecting different geological units. Lower radon concentrations were recorded in the thermal swimming pool of TS9 and *Vichy* shower of TS11. In spite of these thermal spas being located in a geological setting favourable to the radon generation these have mechanical ventilation which may be the main reason for the lower values of radon concentration.

3.3. Indoor gamma dose rates

The indoor gamma dose rate was measured every hour (μSv/h) in different areas of 13 thermal spas: Medical Office (MO), Technical Area (TA), ORL, Thermal Pool (TP) and Steam Area (SA).

The dose rates were registered for periods between 25 and 45 days. The basic statistic parameters are presented in Fig. 4 for average (AV), median (MD), standard deviation (SD), minimum (Min) and maximum (Max).

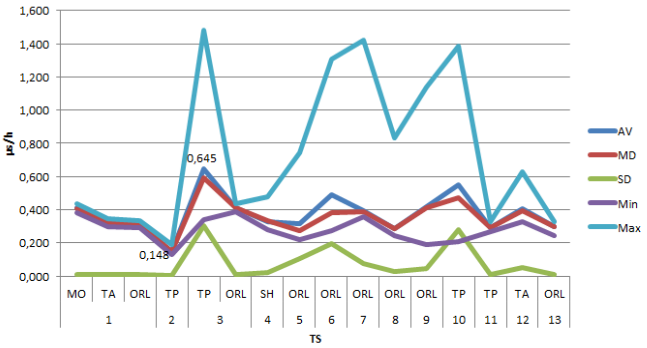

Figure 4. Indoor gamma dose rates (μSv/h)

The average of gamma dose rates ranged between 0,148 μSv/h (TS2) and 0,645 μSv/h (TS3). This reflects the very different geological settings, in particular for the thermal spa with the highest dose rate (SP3) and for the thermal spa with the lowest dose rate (SP2), which are located in a zone comprised predominantly of granitic rock and in a geological framing comprised mostly of metasedimentary rock, respectively.

The gamma dose rates ranged between 0,15 μSv/h and 0,65 μSv/h. The annual dose rate was estimated varying between 0,30 and 1,29 mSv/y, for 2000 working hours per year, which is far below the effective dose limit for workers (20 mSv/y) (Directive 43/EURATOM). However the great contribution for the annual effective dose will result from radon inhalation which is not included in this estimation.

4. Conclusions

The results showed that the recommended limit for drinking water of 100 Bq/L (2001/928/ EURATOM) was exceeded in 51% of the selected thermal spas. About 80% of the total measurements of indoor radon concentration exceeded the previous EU reference level of 200 Bq/m³ for new buildings and about 63% exceeded the new reference level of 300 Bq/m³ (Directive 2013/59/EURATOM). Therefore, as the recommended limits for radon concentration in water and in indoor air were exceeded, appropriate actions should be taken in order to reduce the hazard to health from radon indoors and the potential resulting occupational exposure.

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