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VARIABILITY OF INDOOR RADON LEVEL ACCUMULATION: A STUDY IN PORTUGUESE THERMAL SPAS

A.S. Silva[1,2], M.L. Dinis [1,2*]

[1]CERENA, FEUP/UPorto - Centre for Natural Resources and the Environment,
Faculty of Engineering, University of Porto, R. Dr. Roberto Frias, 4200-465, Porto, Portugal
[2]PROA/LABIOMEP - Research Laboratory on Prevention of Occupational and Environmental Risks,
Faculty of Engineering, University of Porto, R. Dr. Roberto Frias, 4200-465, Porto, Portugal

Abstract. *The aim of this work was to measure the concentration of the indoor radon in 16 Portuguese thermal spas (38% of the thermal spas in Portugal) and assess its variability within each establishment as well as its contribution to the effective dose. The measurements were performed with CR-39 passive detectors placed at different workplaces within each thermal spa, for an average period of 42 days, in different seasons of the year. The indoor radon concentrations ranged from 68 to 4335 Bq/m^3 with a geometric mean of 437 Bq/m^3 and an arithmetic mean of 702 Bq/m^3. Geological factors that can lead to such behaviour are discussed. The results showed that the EU reference level of 300 Bq/m^3 (Directive 2013/59/EURATOM) was exceeded in several cases. No significant differences were observed among measurements taken during different seasons of the year, however, large differences of radon concentrations in different rooms of the same thermal establishment were noted as well as significant difference when comparing to other thermal establishments. The effective dose resulting from the inhalation of radon ranged between 2 and 32 mSv/y. In 43% of the thermal spas, the effective dose is likely to be higher than 6 mSv/y, which means that the exposure should be managed as a "planned exposure situation" according to the European Directive 2013/59/EURATOM. Also, in 19% of the cases, the annual effective dose exceeds 20 mSv/y, and in these cases, monitoring and radiological protection is required as laid down in the European Directive 2013/59/EURATOM.*

Key words: *Radon, effective dose, geology, thermal spa*

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1. Introduction

Exposure to ionizing radiation of natural origin contributes with 81% to the annual dose received by the population, and, in the case of Portugal, most of the average annual dose to which the population is exposed by natural sources is due to radon inhalation and its radioactive progeny (57%), and terrestrial gamma radiation (18%) [1].

Radon is currently recognized as the leading cause of lung cancer, with the exception of tobacco [2, 3, 4, 5, 6, 7].

The exposure to radon and its decay products occurs predominantly in indoor environments like dwellings and workplaces. Most of the radon in indoor air comes from soil underneath the buildings. Radon can also dissolve and accumulate in water from underground sources such as wells and springs. When water that contains radon is used, radon gas escapes from the water and goes into the indoor air. In general, radon is present in the outdoor air in small amounts and it is quickly diluted to very low concentrations. In confined spaces such as basements and subterranean spaces, radon may be present in higher concentrations, in particular, in cases where ventilation is deficient or non-existent.

The main workplaces where the exposure to radon can be considered as occupational exposure are mines and other underground workplaces such as caves and galleries (where high concentrations of radon are accumulated) and thermal spas (exposure through use and handling of thermal water) [8].

The health hazards from radon are mainly due to the inhalation of airborne activity (unattached and attached) of radon decay products. The most important parameters affecting the inhalation dose resulting from these decay products are aerosol size distribution, unattached fraction, breathing rate, and the depth in tissue of the target cell nuclei [9, 10, 11].

Radon concentration varies considerably depending on different parameters: i) source term (topography, soil characteristics, type of dwelling); ii) environmental parameters (temperature, pressure, humidity, wind speed, weather) and iii) other variables such as routine habits and lifestyle of the population.

For indoor radon, the source terms are represented by: soil beneath the buildings, construction materials and water [11, 12]. The soil type, altitude, and

* mldinis@fe.up.pt

geological formation strongly affect the indoor and outdoor doses [11, 13]. For the same region, the indoor radon concentration may be affected by the variation of the construction materials used for building while other factors remain constant (such as pressure differential, weather and climate, etc.) [11, 14]. At the same time, routine habits and lifestyle of the population may be very different, which makes it extremely difficult to relate all the parameters, quantify and interpret the variability of indoor radon concentration.

In Portugal, the geological settings are mostly comprised of granite rocks with uranium mineralization, which represents an elevated risk for high indoor radon levels, particularly in some specific regions. Therefore, in the national legislation, DL 79/2006 [15], the reference value for the maximum indoor radon concentration within the existing buildings is defined (400 Bq/m3), being monitored mandatorily only in dwellings located in granitic areas [16]. Also, Portugal has a long tradition of hydrotherapy therapeutics with mineral water from the occurrences of natural springs, which outcrop, in many cases, from faulty and joins with uranium mineralizations. Therefore, there is a high probability of significant amounts of radon to be present in mineral waters used for therapeutic treatments.

The European Union, through the Directive 96/29/EURATOM [17], identified hydrotherapy as a professional activity during which workers are exposed to radiation from natural sources. Moreover, in the recently published European Directive 2013/59/EURATOM, it is defined that the reference level for the annual average activity concentration in air should not be higher than 300 Bq/m3, unless it is warranted by national prevailing circumstances, being implicit that, in this case, the Member State submits the information to the European Commission.

In what concerns the effective doses, the Directive 2013/59/EURATOM also refers that, where exposure of workers is likely to exceed the effective dose of 6 mSv/year, it should be managed as a "planned exposure scenario" where dose and dose restriction limits (exposure optimization) apply and case The effective dose is equal to or less than 6 mSv/year, the company is required to maintain the exposure of the workers under observation [16].

The aim of this work was to assess the exposure to indoor radon by measuring the concentration of the indoor radon in 16 Portuguese thermal spas (38% of the thermal spas in Portugal) and evaluate its variability within each establishment as well as its contribution to the effective dose. Geological factors that can affect the indoor radon levels, and consequently the annual effective dose, are discussed.

2. Materials and Methods

2.1. Study area

The Portuguese mainland, located in western Iberia, presents three fundamental units, distinct from the chronological point of view, and from the structure of land. These morpho-structural units are: i) Hesperic Massif; (ii) Meso-Cenozoic Occidental and Meridional Basins and (iii) Tejo and Sado Basins.

The great diversity of the geology characteristic of the country (in the north, abound granite and other plutonic rocks, in the centre and south, schist, limestone, sandstone and quartzite) determines the different physical-chemical composition of Portuguese mineral water and therefore, the occurrences of thermal waters have considerably different characteristics. In general, the geothermal potential is directly related to the tectonic features, which beneficiate the circulation of the fluids.

In what concerns the Hesperic Massif, most of the thermal springs are preferably located along NNE, NE and ENE orientation alignments and associated with granitic and shale rocks, as well as rich quartzite minerals of sulfide mineralizations. These are essentially sulfuric waters, which are poorly mineralized, and some are bicarbonate and gasocarbonic, the latter having high mineralizations. Hyposaline waters seem to be correlated mainly with geological environments where quartzite rocks predominate.

In the Meso-Cenozoic Occidental Basin, the geology is dominated by extensive sedimentary formations. The thermal springs occur alongside the faults that originated from the typhonic valleys in areas of gypsum-saline diapiris. As a result, these waters are chlorate-bicarbonate-sodium, and the presence of sulphate-calcium hot springs is also noteworthy. These are hypersaline waters with pH values close to neutrality.

In the Meso-Cenozoic Meridional Basin, the thermal waters are essentially bicarbonate-sodic and bicarbonate-calcium, due to the interactions with igneous and sedimentary rocks, respectively.

This study was performed in 16 thermal spas (TS) located in the following districts: Aveiro (1 TS), Braga (3 TS), Bragança (1 TS), Castelo Branco (1 TS), Guarda (3 TS), Porto (2 TS), Viana do Castelo (1 TS), and Viseu (4 TS).

These eight districts are mainly located in the North/Center region of Portugal where most of the natural springs, with emergency temperatures between 20 ºC and 76 ºC, are located (Fig. 1).

In what concerns the geological characteristics of the regions where, in particular, the studied thermal spas are located (obtained from the geological maps of Portugal), 69% (11/16) of the TS are located mostly in granitic regions, 25% (4/16) in regions with granite and schist substrate and 6% (1/16) in a region of metasediment rocks (Table 1).

For lithology, and in particular in what concerns uranium and thorium mineralizations, only one Portuguese geological map (Viseu 17A - where TS6 and TS3 are located) presents data for these mineral occurrences. For the local region where TS6 is located, ^{40}K, ^{235}U and ^{232}Th occur with concentrations of 40800±20.4 ppm; 1260±1.2 ppm, and 7.4±1.2 ppm, respectively [22], while for the local region where TS3 is located, these values are: 46600±1530 ppm, 118±7.1 ppm and 9.4±1.0 ppm, respectively.

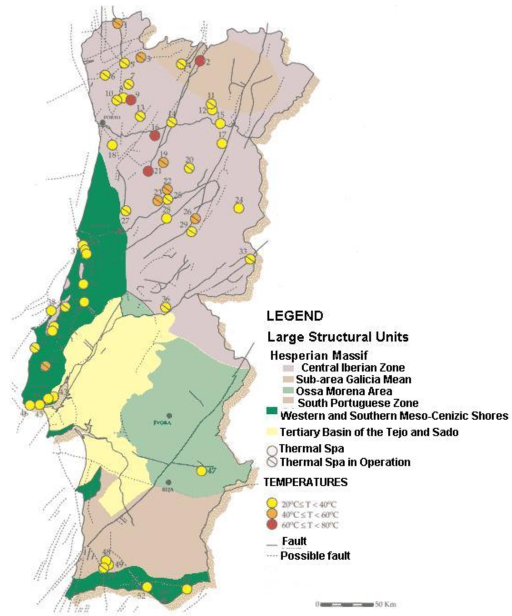

Figure 1. Thermal spas in Portugal

Table 1. Geological structure of the regions where the studied thermal spas are located

TS	Geological map (1:50000)	Geological information	Structure of the Massif	Type of ventilation
TS1	16B [18]	Metasediment Rocks	Fault Intersection	NV/MV
TS2	17A [19]	Granite and Schist Substrate	Penacova-Régua-Verin	NV/MV
TS3	17C [20]	Granite Substrate	--	NV/MV
TS4	11C [21]	Granite and Schist Substrate	Fracturing dependency	NV
TS5	17A [19]	Granite and Schist Substrate	Rio Dão Fault	NV/MV
TS6	5B [22]	Granite Substrate	Fault Intersection	NV/MV
TS7	5B [22]	Granite Substrate	Fault Intersection	NV
TS8	13B [23]	Granite Substrate	Fault Intersection	NV
TS9	20B [24]	Granite Substrate	Fault Intersection	NV
TS10	18C [25]	Granite Substrate	--	NV/MV
TS11	20B [24]	Granite Substrate	Fault Intersection	NV
TS12	17C [20]	Granite substrate	Fault Intersection	NV/MV
TS13	15A [26]	Granite Substrate	Vilariça - Manteigas Fault	NV/MV
TS14	9B [27]	Granite Substrate	Fault Intersection	NV/MV
TS15	5D [28]	Granite Substrate	Fault Intersection	NV
TS16	1B [29]	Granite and Schist Substrate	Rio Minho Fault	NV

For the radiogenic potential of the region where TS6 is located, the concentrations of U and Th are 7.4±1.2 ppm and 20.4±1.2 ppm, respectively, while for TS3, the concentrations of U and Th are 8.5±1.4 ppm and 12.1±3.9 ppm, respectively. The geological map of Viseu 17A also shows that the radiogenic potential for the indoor air is considered high for both locations [22].

2.2. Sampling plan

The study was conducted during two sampling campaigns for radon concentration measurements and dose assessment. The first campaign was carried out between November 2013 and June 2014 and the second campaign took place between July 2014 and September 2015.

In some thermal spas (TS4, TS5, TS6, TS7) the measurements were carried out in a campaign, since they are open to the public for 4 months. In the case of the TS16 thermal spa, the measurements were only made in a campaign because the thermal spa closed to the public due to works.

Radon concentration in the indoor air of the thermal spas was measured in different spaces of permanence of workers: access corridor to the thermal pool (AC), buvete hall (BH), bathtubs (BT), hall spa (HS), jet shower (JS), ORL (inhalation techniques), treatment area (TA), thermal pool (TP), vapors (VP), vichy shower (VS), machines house (MH) in different seasons (spring/summer and autumn/winter) and, on average for a period of 42 days.

In order to proceed to the identification and characterization of the thermal spas, a questionnaire was filled during the first visit to the locality. The questionnaire is divided into 2 sections: i) identification and characterization of the conditions of the operation of the thermal spas and (ii) characterization of the installations.

The first section deals with issues related to work schedules, gender, seniority in the work place and number of thermalists per year.

The second section presents the criteria used for the characterization of thermal spa facilities, namely the existence of risk-assessment procedures, previous studies on the measurement of radon concentrations, the existence or nonexistence of a radiological control plan (RCP), safety services and health at work (SSHW), type of ventilation (natural ventilation (window opening) mechanical ventilation), presence of air conditioning, existence of sludge treatment and thermal pool. A survey for the description of the characteristics of the equipment used in therapeutic treatments was also included.

2.3. Measurement methods

The measurements of radon concentrations in the indoor air were performed using CR-39 nuclear track detectors (5-cm height, 3-cm diameter) enclosed in small cylindrical diffusion chambers (Fig. 2). These detectors comprise a small piece of polycarbonate, highly sensitive to ionizing particle tracks such as alpha particles [30].

The CR-39 detectors were placed approximately 2 meters from the floor. At the end of each time period (on average 42 days), the detectors were removed and stored individually in sealed containers to prevent any contamination from other sources during transport to the laboratory [31]. The analysis was performed in the Natural Radioactivity Laboratory in the Department of Earth Sciences of the University of Coimbra.

The Laboratory of Natural Radioactivity of the Department of Earth Sciences of the University of

Coimbra regularly participates in comparison exercises with other laboratories in order to estimate the statistical uncertainty (analytical error of less than 10% of the value obtained). The limit of detection using the described procedure is 5 Bq/m^3.

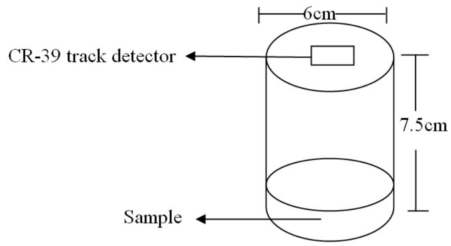

Figure 2. CR-39 nuclear track detectors

The inhalation dose (D, mSv/y) was calculated from the results obtained for the indoor radon concentration (*CRn*, Bq/m^3) and considering the following exposure parameters in indoor environments (ICRP, 1994; UNSCEAR, 2000) (Eq. 1): for the exposure time (ET), an occupancy of 2000 h/y for the exposure within the thermal spas, an equilibrium factor between radon and its progeny of 0.4 and a dose conversion factor of 9×10^{-6} (mSv/y per Bq/m^3) (effective dose received by adults per unit of ^{222}Rn activity per unit of air volume).

$$D = CRn \times 0.4 \times ET \times 9 \times 10^{-6} \qquad (1)$$

Although there is job rotation in all the thermal spas (with the exception of TS8), the dose was assessed considering the "worst-case scenario", assuming that workers do not have job rotation. Therefore, the dose was calculated with the most conservative value of each input [30], meaning that a hypothetical situation was considered in which everything that contributes to the exposure was maximized, namely the exposure time (2000 h/y) and indoor radon concentration (maximum value) [32].

The dose assessment was considered only for workers whose exposure will be reflected in a long-term impact (longer and continuous exposure) while patients or spa users will experience a short-term impact, probably once in a lifetime and negligible.

3. Results and discussion

3.1. Indoor radon concentration

Radon concentration values in thermal spas ranged from 73 Bq/m^3 to 4335 Bq/m^3. The highest value was obtained in TS4 and the minimal value was obtained in TS11 (Table 2).

By analyzing the Table 3, the values of radon concentration in the indoor air of the thermal spas TS1, TS2 (winter), TS3, TS4, TS5, TS6, TS7, TS10 (winter and autumn), TS12, TS14 (winter), TS15 and TS16 exceed the 300 Bq/m^3 projected in the European Directive 2013/59/EURATOM. In only 25% of thermal spas, the radon concentration in indoor air is less than 300 Bq/m^3: TS8, TS9, TS11 and TS14.

In the thermal spas TS1, TS2, TS10, TS12, TS15 and TS16, the values obtained for the radon concentration in the indoor air in winter/autumn and spring/summer seasons are due to poor ventilation on the premises and it is assumed that radon predominantly originates from the subsoil, as these thermal spas are located in regions with granite substrate.

On the other hand, in TS3, the values of the indoor radon concentration, obtained in spring and summer, are very similar.

In TS4 it can be observed that radon concentration in indoor air is quite high, which is explained by the geological conditions, since this thermal spa is located in a predominantly granitic zone.

However, in TS5 it is assumed that the strong contribution of radon is from the thermal water, since the values of radon concentration in the water are quite high, and since in these places mineral water is used for therapeutic treatments.

Within TS6, the great difference in the values obtained for radon levels in the existing treatment rooms is due to the insufficient ventilation in some places, namely in the JS.

Table 2. Indoor radon concentration (Bq/m3) in the studied thermal spa

TS	Location	WI		SP		SU		AU	
		^{222}Rn	AV	^{222}Rn	AV	^{222}Rn	AV	^{222}Rn	AV
TS1	BT	674	1646	--	--	436	1296	--	--
	ORL	3479		--		3119		--	
	TP	784		--		333		--	
TS2	AC	566	566	--	--	--	237	--	
	ORL	329		--		187		--	
	TA	692		--		--		--	
	TP	517		--		267		--	
	VS	724		--		258		--	
TS3	ORL	--	--	502	413	489	442	--	--
	TA	--		401		429		--	
	TP	--		274		333		--	
	VP	--		453		465		--	
	VS	--		437		495		--	
TS4	ORL	--	--	--	--	4335	3124	--	--
	VS	--		--		1912		--	
TS5	ORL	--	--	--	--	1190	1256	--	--
	RB	--		--		953		--	
	SH	--		--		878		--	
	TP	--		--		2181		--	
	VP	--		--		1173		--	
	VS	--		--		1163		--	
TS6	BH	--	--	--	--	1615	1047	--	--
	JS	--		--		1681		--	
	ORL	--		--		366		--	
	TP	--		--		423		--	
	VS	--		--		1148		--	
TS7	ORL	--	--	--	--	347	354	--	--
	VS	--		--		361		--	
TS8	ORL	--	--	169	273	--	--	143	252
	VS	--		376		--		360	
TS9	ORL	--	--	169	232	--	--	269	234
	TP	--		121		--		204	
	VS	--		406		--		229	
TS10	AC	641	705	--	255	--		209	312
	LP	1079		--		--		377	
	ORL	--		255		--		--	
	TA	481		--		--		305	
	TP	618		--		--		358	
TS11	HS	--	--	116	153	--	--	132	222
	ORL	--		312		--		498	
	TP	--		73		--		101	
	VS	--		112		--		155	
TS12	JS	--		1130	1608	--	--	--	2441
	ORL	--		2298		--		1643	
	TA	--		1145		--		--	
	TP	--		1494		--		2808	
	VS	--		1971		--		2873	
TS13	ORL	--	--	146	147	--	--	235	184
	TP	--		203		--		176	
	VS	--		93		--		141	
TS14	BT	172	356	--	--	266	219	--	--
	ORL	375		--		175		--	
	SA	467		--		214		--	
	TP	370		--		240		--	
	VP	398		--		199		--	
TS15	ORL	707	580	--	--	--	--	--	--
	TP	355		--		--		--	
	AC	841		--		--		--	
	MH	422		--		--		--	
TS16	MH	1692	1122	--		--		--	
	TP	862		--		--		--	

Av – average arithmetic; Max – maximum; Min - Minimum

In TS7, TS8 and TS9, TS11, TS13, TS14 the values obtained in the different locations are very similar and close to the limit required by the legislation.

On the other hand, it is in the ORL that the values of radon concentration in the indoor air of the thermal spas tend to be high (Table 2 and Fig. 3). However, the values of radon concentration in the indoor air tend to be lower in the AC (figure 3).

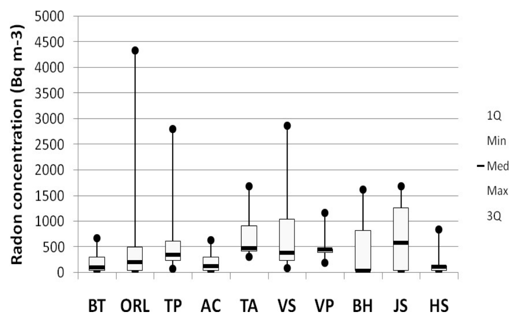

Figure 3. Concentration of radon in indoor air by location

It is also verified that radon levels are significantly higher in the winter months, since due to the heating of the rooms, the hot air rises, creating a negative pressure in the lower floors and this thermal effect leads to the suction of radon from the ground to the building and also because spaces are less ventilated during the winter (Fig. 4).

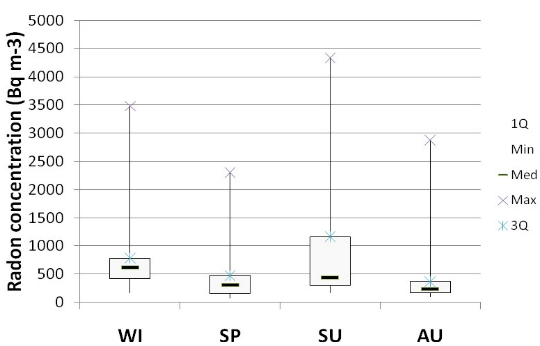

Figure 4. Radon concentration in indoor air *versus* seasons of the year

3.2. Dose assessment

The values of the annual effective dose ranged between 1.21 mSv/y and 31.21 mSv/y. In general, the annual effective dose is below 6 mSv/y, with the exception of TS1, TS4, TS5 (ORL), TS10 (LP) and TS12 (TP) (Fig. 5).

The highest value was obtained in TS4 in the ORL, while the lowest value was obtained in TS8 also in the ORL.

According to the new Directive 2013/59/Euratom, workers from thermal spa establishments are facing a planned exposure situation where dose limits apply. In all other cases, where the effective dose is equal to or less than 6 mSv/y, it is required to keep the exposure under observation.

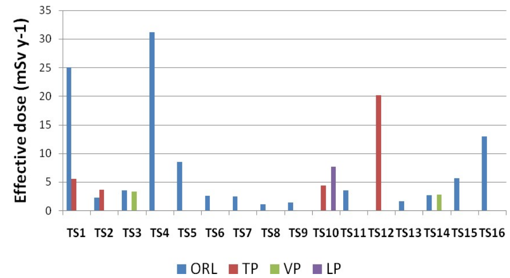

Figure 5. Effective dose by thermal spa

4. Conclusion

In 88% of the results, the indoor radon concentration is above the reference level recommended by the EU. The main reason for these results is due to the geological setting where the thermal spas are located, namely in a granitic region.

Despite the geological (predominantly granitic) condition, in 18% of thermal spas the levels of radon concentration in indoor air were below the reference levels recommended by the EU. This may be justified by the effective ventilation system inside the thermal spa (mechanical ventilation system). Generally, the main source of the radon is the soil but in some cases the construction materials can be also the main source.

In most thermal spas, the highest results of radon concentration in indoor air were obtained during the winter period because the spaces are less ventilated during this period.

It can be seen that in 50% of the thermal spas (TS2, TS4, TS5, Ts10, TS11, TS12, TS15 and TS16) there is a great variability of the values of the radon concentration in indoor air. This is due to the ventilation conditions of the different spaces and it is assumed that radon originates from the subsoil and/or building materials due to the geological setting where these thermal spas are located.

In 31% of the thermal spas, the annual effective dose is higher than 6 mSv/y, which is the reason why it is necessary to maintain the effective annual dose of the workers under observation. However, in 39% of thermal spas, workers face a situation of planned exposure.

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