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Indoor radon levels and total gamma dose rates measurements in Portuguese thermal spas

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Abstract

In this study, an assessment of indoor radon concentration and gamma dose rates were carried out in 16 Portuguese thermal spas. Indoor gamma dose rates measurements were made using a Geiger counter (Gamma Scout – GS3) and radon concentration measurements were carried out using CR-39 detectors. The detectors were exposed for an extended period of time (in average 42 days). The results showed that gamma doses rates are generally low but, in several cases, radon concentration exceeded national as well as international standards, namely the reference level recommended by the Directive 2013/59/EURATOM (300 Bq/m3) and the threshold for protection (400 Bq/m3) from the Portuguese legislation. The annual effective doses deriving from external radiation (gamma dose rates) and indoor radon concentration showed the need to implement measures to optimize the radiation protection of the workers against ionizing radiation.

1. INTRODUCTION

The exposure to natural sources of ionizing radiation is the most significant contribution to the annual dose received by the general population. On average, 80% of the annual dose is due to naturally occurring terrestrial and cosmic radiation sources. The largest natural source of radiation to human exposure is radon gas and its progeny which can contribute with more than 50% of the total dose from natural sources (UNSCEAR, 2000).

Radon is generated from the radioactive decay of radium, an element of the radioactive decay chain of the Uranium-238. In general, uranium occurs naturally at different levels in rocks and soils. Part of the radon produced in rocks and soils escapes to the air; therefore radon is present in the atmosphere where people everywhere may be exposed to radiation from radon itself and also from short-lived radon decay products (Dinis and Fiúza, 2005; UNSCEAR, 2006).

Radon is also soluble in water, and groundwater passing through uranium-bearing soils and uranium-rich rocks (e.g. uranium-rich granites and pegmatites) commonly has significant radon concentrations. In these cases, if groundwater is used as drinking water, people are exposed both through water consumption and also by the inhalation of radon released from the water to the air (UNSCEAR, 2006).

While radon and its daughters continually decay inside the body affecting mostly the lungs, radon daughters ingested with water can cause damage to others organs such as the stomach tissue. Some radon and its progeny swallowed in drinking water pass through the stomach walls and intestine (ATSDR, 2010). Nevertheless, the risk of radon exposure is mostly associated with high radon concentrations in confined environments and the subsequent inhalation (Silva and Dinis, 2016).

Radon gas has been identified as a leading cause of lung cancer, second only to cigarette smoking (UNSCEAR, 2000, 2006; DGS, 2002; IAEA, 2003; WHO, 2007, 2009). Radon gas is responsible for an estimated 21 000 deaths from lung cancer annually and 2 900 of these deaths occur among people who have never smoked (EPA, 2009). The risk of cancer due to radon exposure is increased for smokers, as the radiation emitted by tobacco synergizes when in the presence of radon gas (Darby, 2005; Al Zoughool et al., 2009; EPA, 2013; Erdogan et al., 2013).

It is recommended that indoor radon concentration be reduced to levels below than 100 Bq/m^3 (annual average) or, if this is not possible, that in a first step, a limit of 300 Bq/m^3 should be adopted being then progressively reduced to 100 Bq/m^3 (WHO, 2009). On the other hand, the EU recently approved as a reference radon concentration to limit the exposure to radon, the value of 300 Bq/m^3 (annual average) in the indoor air of buildings including homes and workplaces. This benchmark has changed the previous recommendations (400 and 200 Bq/m^3 for existing buildings and new constructions, respectively) and should now be implemented in all Member States (Directive 2013/59/EURATOM, 2013). In Portugal, the Decreto-Lei 118/2013 (DL, 2013), states that the protection thresholds for concentrations of indoor air pollutants, including radon, are set out in Portaria 353-A/2013, specifying the threshold of 400 Bq/m^3 for radon exposure, being its measurement and monitoring mandatory only in buildings built in granitic zones, namely in Braga, Castelo Branco, Guarda, Porto, Vila Real and Viseu districts.

Concerning dose limits, the Directive 2013/59/EURATOM states that where exposure of workers is likely to exceed the effective dose of 6 mSv/year, it should be managed as a "planned exposure situation" where there apply dose limits (exposure optimization) and if the effective dose is less than or equal to 6 mSv/year, then it is required that workers' exposure should be kept under surveillance.

The use of thermal mineral water in the treatment of diseases, known as hydro- and hydrothermal therapy is one of the professional sectors with potential for exposure to natural radiation sources in large part due to the inhalation of radon released from thermal waters (Silva and Dinis, 2016).

Portugal is a country with some risk in relation to natural radiation, since in many regions of the country the soil is composed by granitic rocks and these may contain extensive uranium mineralizations (ITN, 2010; Silva et al., 2014; Silva and Dinis, 2016). Most of the thermal springs existing in the Portuguese territory (with emergency temperatures between 20 ºC and 76 ºC) are located in the North/Center of the country due to the geological and structural characteristics of these regions (Figure 1).

Several studies have been carried out in different countries: Algeria (Ziane et al., 2014), Austria (Bossew et al., 2007), Brazil (Alberigi et al., 2011), China (Song et al., 2011), Croatia (Radolić et al., 2005), Greece (Nikolopoulos et al., 2010), Hungary (Hámori et al., 2006), Serbia (Nikolov et al., 2012), Spain (Ródenas et al., 2008), Poland (Walczac et al., 2016) and Turkey (Tarim et al., 2012) to measure the radon concentration in indoor air and in natural mineral water of thermal spas in order to estimate the doses of natural radiation received by the workers. In the majority of these studies and due to several reasons, radon concentration can reach worrying levels. In one of these studies, carried out in the southeastern part of Serbia, where there are several occurrences of hot springs from igneous and metamorphic rocks, a correlation was established between the occurrence of high concentrations of radon in natural mineral waters and geological and hydrogeological factors (Koray et al., 2014). There are also records of very high radon levels in some thermal hotels in China, representing an effective risk to workers (Song et al., 2011; Abbasi et al., 2013).

Although in Portugal there is a long tradition in thermal water therapy, very few studies have been conducted addressing the problem of the radiation exposure due to radon inhalation, in particular for workers and none of these was conducted by the regulatory bodies. Therefore, the present study contributes to the knowledge and the understanding of the situation in what concerns to the radon exposure within the Portuguese thermal spas, where an assessment of indoor radon concentration and gamma dose rates was carried in order to assess the occupational exposure incurred for workers from this sector, detailed in Silva and Dinis (2016).

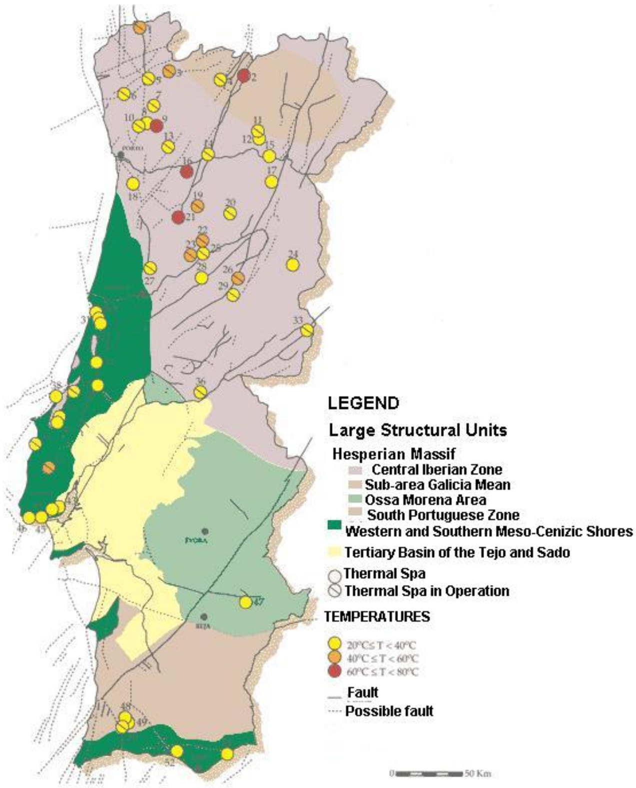

Figure 1. Thermal spas in Portugal.

2. MATERIALS AND METHODS

A preliminary evaluation of radiological hazards of a standard MSW incinerator facility is made on theoretical grounds with the purpose to identify and generically assess radiation hazards associated to such facility.

This study was carried out for 3 years in 16 thermal spas from a total of 38 (42% of the existing thermal spas) that accepted to participate in the study. The study is a follow-up of a previous (Silva and Dinis, 2016), that comprised periodically measurements of indoor radon concentration and gamma dose rates. Effective dose was then calculated with the collected data.

Indoor radon concentration and gamma dose rates measurements were taken between November 2013 and September 2015 during two different periods: spring/summer (SP/SU) and autumn/winter (AU/WI). These locations were identified as: AC (access corridor to the thermal pool), BH (buvete hall), BT (bathtubs), HS (hall spa), JS (jet shower), LP (ludic pool), ORL (inhaler techniques room), RB (rest balcony), SA (sludge area), SH (steam hall), TA (treatment area), TP (thermal pool), VP (vapors), VS (vichy shower), MH (machines house), MO (medical office) and RP (rehabilitation pool). Measurements of gamma radiation doses were carried out in some of the previous locations where employees spend most of their time (Silva and Dinis, 2016; Silva et al., 2016).

The measurements of radon concentrations in air were performed using CR-39 detectors (detection limit of 0.01 Bq/m^3) enclosed in small cylindrical (5-cm height, 3-cm diameter) diffusion chambers for periods between 25 and 45 days. The CR-39 detectors were placed in each room at approximately 2 meters from the floor. After the period of exposure (in average

42 days), the detectors were retrieved and sent to the Natural Radioactivity Laboratory of the Department of Earth Sciences, University of Coimbra, Portugal (Silva and Dinis, 2016).

The detectors were etched in 25% NaOH solution at 90ºC for 270 minutes. The number of tracks in an area of 1 cm^2 on each film was counted by a microscope automatic reader. The background track density was then subtracted and related to radon concentration level using a calibration factor obtained by the exposure of detectors of the same batch in a certified calibration chamber.

Indoor gamma dose rate measurements were carried out using a dose rate meter (1.20 meters above the floor) (Gamma Scout Geiger Counter) which is a calibrated measurement instrument for alpha, beta and gamma rays. The registers were hourly acquired and stored for approximately 45 days (Silva et al. 2015; Silva and Dinis, 2016).

The inhalation dose (D, mSv/y) was calculated from the results obtained for the indoor radon concentration (CRn, Bq/m^3) and for the following exposure parameters in indoor environments (ICRP, 1994; UNSCEAR, 2000): an occupancy of 2000 h/y, an equilibrium factor between radon and its progeny of 0.4 and a dose conversion factor of 9 x 10^{-6} Sv (Bq h m^{-3}) (ICRP, 1994; UNSCEAR, 2000):

$$D\,(\text{mSv/y}) = C_{Rn} \times 0.4 \times 2000 \times 9 \times 10^{-6} \quad (1)$$

For the assessment of the effective dose, due to external exposure in indoor environments, the same exposure parameters (occupancy 2000 h/y) were used combined with the data from the dose meters (Gamma Dose Rates - GDR, mSv/h):

$$D\left(\text{mSv/y}\right) = GDR \times 2000 \quad (2)$$

3. RESULTS AND DISCUSSION

3.1. Radon concentration in the indoor air

Table 1. Indoor radon concentration in the studied thermal spas (TS).

TS	SEASON	^{222}Rn (Bq/m3)															
		BT	ORL	RB	SH	TP	AC	VS	VP	SA	JS	LP	HS	TA	MH	MO	RP
1	WI	674	3479	--	--	784	--	--	--	--	--	--	--	--	--	--	--
	SU	436	3119	--	--	333	--	--	--	--	--	--	--	--	--	--	--
2	WI	--	329	--	--	517	566	724	--	--	--	--	--	692	--	--	--
	SU	--	187	--	--	267	--	258	--	--	--	--	--	--	--	--	--
3	WI	--	502	--	--	274	--	437	453	--	--	--	--	401	--	--	--
	SU	--	489	--	--	333	--	495	465	--	--	--	--	429	--	--	--
4	SU	--	4335	--	--	--	--	1912	--	--	--	--	--	--	--	--	--
5	SU	--	1190	953	878	2181	--	1163	1173	--	--	--	--	--	--	--	--
6	SU	1615	366	--	--	423	--	1148	--	--	1681	--	--	--	--	--	--
7	SU	--	347	--	--	--	--	361	--	--	--	--	--	--	--	--	--
8	SP	--	169	--	--	--	--	376	--	--	--	--	--	--	--	--	--
	AU	--	143	--	--	--	--	360	--	--	--	--	--	--	--	--	--
9	SP	--	169	--	--	121	--	406	--	--	--	--	--	--	--	--	--
	AU	--	269	--	--	204	--	229	--	--	--	--	--	--	--	--	--
10	WI	--	--	--	--	618	641	--	--	--	--	1079	--	481	--	--	--
	SP	--	255	--	--	--	--	--	--	--	--	--	--	--	--	--	--
	AU	--	--	--	--	358	209	--	--	--	--	377	--	305	--	--	--
11	SP	--	312	--	--	73	--	112	--	--	--	--	116	--	--	--	--
	AU	--	498	--	--	101	--	155	--	--	--	--	132	--	--	--	--
12	SP	--	2298	--	--	1494	--	1971	--	--	1130	--	--	1145	--	--	--
	AU	--	1643	--	--	2808	--	2873	--	--	--	--	--	--	--	--	--
13	SP	--	146	--	--	203	--	93	--	--	--	--	--	--	--	--	--
	AU	--	235	--	--	176	--	141	--	--	--	--	--	--	--	--	--
14	WI	172	375	--	--	370	--	--	398	467	--	--	--	--	--	--	--
	SU	266	175	--	--	240	--	--	199	214	--	--	--	--	--	--	--
15	WI	--	707	--	--	355	--	--	--	--	--	--	841	--	422	577	--
16	WI	--	--	--	--	862	--	--	--	--	--	--	--	--	1692	--	813

The values obtained for the indoor radon concentration in each one of the selected thermal spas (TS) and the respective location are presented in the Table 1 (the measurement of the radon concentration was carried out according to where employees spend most of their time). The season when the measurements were taken is also identified: SP – spring; SU – summer; AU – autumn WI – winter.

The average radon concentration in indoor air is 716 Bq/m^3. The lowest value was obtained at TS11 during spring (73 Bq/m^3) and the highest value was obtained at TS4 (4335 Bq/m^3) during summer. It was not possible to observe a seasonal effect between the different measurements periods of radon concentration. Also, in average, the radon concentration was higher than both the reference level recommended by the Directive 2013/59/Euratom (300 Bq/m^3) and the threshold for protection, laid down in the Portuguese legislation (400 Bq/m^3) (Portaria nº 353-A/2013; Portaria, 2013).

The lowest values of indoor radon refer to the swimming pools which may be justified by the fact that this water is not daily replaced. The highest values were obtained in the ORL (inhalation therapy rooms). Although these rooms have natural ventilation it seems to be inefficient in decreasing the indoor radon levels (Silva and Dinis, 2016).

The considerable variability of the radon concentration observed in each thermal establishment is influenced by many factors, namely: rock type, double glazing, window opening habits, building materials used in the walls, type of flooring and draught proofing, the tightness of the building barriers with the soil and, ultimately, on the ventilation level, natural or mechanical (Faísca et al., 1992; Gunby et al., 1993).

3.2. Gamma Dose Rates

The measurements of gamma dose rates occurred simultaneously with the measurements of the indoor radon concentration at the following locations: ORL (inhaler techniques room), TP (thermal pool) and SA (sludge area). The results are presented in Table 2 including the number of hourly readings (Silva and Dinis, 2016).

Table 2. Gamma dose rates measurements in the studied thermal spas (TS).

TS	Location	Gamma Dose Rates (µSv/h)				
		Average	Standard deviation	Maximum	Minimum	Medium
TS1	ORL	0.306	0.226	1.282	0.132	0.201
TS2	SA	0.347	0.045	0.434	0.277	0.323
TS3	TP	0.484	0.274	1.484	0.227	0.396
TS4	SA	0.291	0.037	0.478	0.067	0.292
TS5	ORL	0.318	0.105	0.741	0.218	0.274
TS6	ORL	0.490	0.197	1.306	0.271	0.381
TS7	ORL	0.393	0.020	1.423	0.235	0.387
TS8	ORL	0.285	0.025	0.831	0.246	0.284
TS9	ORL	0.417	0.046	1.137	0.190	0.412
TS10	TP	0.423	0.242	1.387	0.206	0.291
TS11	TP	0.290	0.040	0.352	0.197	0.304
TS12	SA	0.406	0.053	0.630	0.330	0.392
TS13	ORL	0.293	0.009	0.327	0.242	0.293
TS14	TP	0.339	0.023	0.419	0.018	0.338
TS15	ORL	0.419	0.205	1.036	0.279	0.307
TS16	SA	0.295	0.009	0.321	0.259	0.295

The gamma dose rates ranged between 0.018 and 1.484 µSv/h. In average, the highest gamma dose rate was obtained in inhaler techniques rooms (0.490 µSv/h) following the thermal pool (0.484 µSv/h and 0.423 µSv/h). Both maximum of gamma dose rates and minimum values were obtained in the thermal pool. The values obtained are associated with the construction materials of the thermal spas. In average, the reference level the European Directive 2013/59/EURATOM for members of the public (1 mSv per year) was exceeded in all monitored locations, 3.17 mSv/y.

3.3. Assessment of the annual effective doses

The dose was assessed considering the "worst-scenario" assuming that workers do not have job rotation and maximum radon concentration in indoor air. The annual effective doses due to radon inhalation were calculated in the different thermal spas (Table 3).

Table 3. Annual effective dose in the studied thermal spas (TS).

TS	Location	Effective dose (mSv/y)
1	ORL	25.05
2	SA	2.37
3	TP	3.35
4	SA	31.21
5	ORL	8.57
6	ORL	2.63
7	ORL	2.50
8	ORL	1.21
9	ORL	1.47
10	TP	4.45
11	TP	3.59
12	SA	20.21
13	ORL	1.69
14	TP	2.70
15	ORL	12.18
16	SA	5.09

The effective dose of ionizing radiation due to indoor radon was calculated according to equation 1 and the results ranged from 1.09 to 31.21 mSv/y, within the thermal spas.

In new Directive 2013/59/EURATOM, the Euratom Treaty defines the "basic standards" for the protection of the health of workers and the general public against the dangers arising from ionizing radiations. It is stated that "*where radon enters from the ground into indoor workplaces, this should be considered to be an existing exposure situation since the presence of radon is largely independent of the human activities carried out within the workplace. Such exposures may be significant in certain areas or specific types of workplaces to be identified by Member States, and appropriate radon and exposure reduction measures should be taken if the national reference level is exceeded*".

In what concerns to radon in workplaces if the concentration remains above the national reference level (despite optimization) it is necessary to notify the competent authority and introduce occupational exposure arrangements. Above an annual effective dose of 6 mSv/y, the situation is to be managed as a planned exposure situation (and dose constraints or reference levels of 1–20 mSv are to be applied, requiring exposure optimization) and equal or below 6 mSv/y the exposure needs to be kept under review. All Member States have to transpose the new Directive to national legislations until February of 2018; until then, the previous Directive 96/29/EURATOM shall apply.

4. CONCLUSIONS

The indoor radon concentrations observed in some of the thermal spas were elevated and are the main contributors to the total annual effective dose of workers. The gamma doses (external exposure) were in general low, meaning a relatively low contribution to the total annual dose. However, in several cases, radon concentration exceeded the national as well as the international standards: reference level recommended by Directive 2013/59/EURATOM (300 Bq/m^3) and the threshold for protection (400 Bq/m^3) from the Portuguese legislation. 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. In these cases, the explanation for this fact is due to the effective ventilation system inside the thermal establishment (mechanical ventilation system). In most thermal establishments the highest indoor radon concentrations were obtained during the winter period, which is consistent with the literature (Ziane et al., 2014) consulted and other studies developed, in which the indoor radon concentration in autumn/winter is higher than the radon concentration in indoor air in spring/summer. The considerable variability of the radon concentration observed in each thermal establishment is influenced by many factors, namely: rock type, double glazing, window opening habits, building materials used in the walls, type of flooring and draught proofing (Madureira et al., 2016).

The annual effective doses deriving of radon concentration in indoor air showed the need to implement measures to optimize the radiation protection of the workers against ionizing radiation. This issue should be adequately addressed to minimize the worker's exposure to ionizing radiation in thermal spas.

The results obtained in this preliminary study carried out in Portuguese thermal establishments concluded that it is necessary to continue the work to evaluate the occupational exposure to radon in a larger sample. It is also important to extend the data collection period over a period of up to one year continuous, due to the high fluctuations of the radon concentration over time.

5. LIMITATIONS

The time horizon in which the study was developed did not allow to identify and study the various variables that influence the radon concentration in the indoor air of the thermal establishments.

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