Radiological Characterization of Selected Thermal Centres Based on Indoor Radon Dosimetry – 17507

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ABSTRACT

In countries with long historical tradition in thermal water therapy and with several mineral-medicinal thermal sources, workplaces within thermal centers may be a source of radon exposure which may be intensified if these are located in regions of high level of natural radiation.

According to the EU Directive 96/29/Euratom (repealed by the Directive 2013/59/EURATOM) each Member State shall identify, by means of survey or any other adequate mean, the work activities where a significant increase in the exposure from natural radiation sources may occur, including thermal centers where the exposure to thoron/radon daughters or gamma radiation may occur.

The purpose of this work was to perform a radiological characterization of selected thermal centers based on indoor dosimetry. The effective doses received by workers due to radon inhalation were estimated and gamma dose rates were measured through continuous periods. The radon risk for indoor exposure was assessed on a probabilistic basis both on thermal centers and workers' dwellings. Radon levels within the thermal centers ranged from 73 to 4335 Bq/m^3 and the values within the workers dwellings are of the same order of magnitude, 68-4051 Bq/m^3. Approximately 66 % of indoor radon concentration values are above the maximum EU reference (300 Bq/m^3) and 94 % of the effective dose is higher than 1 mSv/year. In some situations, radon levels at residential environments are much higher than at workplaces and effective doses are higher than 6 mSv/year, both at residential and work environments.

INTRODUCTION

Radon-222 is a naturally occurring radioactive gas produced by the alpha decay of radium-226 which is present in uranium ores, phosphate rocks, igneous and metamorphic rocks such as granite, gneiss and schist. Radon can in turn disintegrate, producing polonium, bismuth and lead, which are also radioactive, attach themselves to airborne particles and accumulate in enclosed spaces such as basements. The risk of radon exposure is mostly associated with high radon concentrations in confined environments and the subsequent inhalation. Most inhaled radon is rapidly exhaled, but the inhaled decay products readily deposit in the lung epithelium irradiating sensitive cells and thereby enhance the risk of lung cancer.

The exposure to radon gas and its progeny contribute with more than 50% of the total dose from natural sources and it is recognized as the most important cause of lung cancer incidence except for smoking [1]. Thus, the assessment of indoor radon levels is important from the point of view of radiological protection and public health [2], [3], [4].

The World Health Organization (WHO) recently recommended that the concentration of indoor radon should not exceed 100 Bq/m³ or 300 Bq/m³ in exceptions cases, if the above indicated cannot be achieved [3]. The International Commission on Radiological Protection (ICRP) has revised in the latest Statement on Radon the reference level for radon gas in dwellings and other buildings with high occupancy rates to the public by 300 Bq/m³ [5].

In the Directive 96/29/EURATOM, the EU has proposed spa therapy as a professional activity of enhanced natural radiation exposure due, in large part, to the inhalation of radon released from thermal waters. The same Directive (repealed by Directive 2013/59/EURATOM) refers that each Member State shall identify, by means of survey or any other adequate mean, the work activities where a significant increase in the exposure due to natural radiation sources may occur. These include, in particular, thermal centres where thoron/radon daughters or gamma radiation may be present [6].

In countries with long historical tradition in thermal water therapy and with several mineral-medicinal thermal sources, the potential to radon exposure may be intensified if these are located in regions of high level of natural radiation. 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 very high uranium content among others. In these regions it is expected very high indoor radon concentration [7]. Moreover, Portugal is one of the European countries with long historical tradition in thermal water therapy with a total of 38 active thermal centres (Fig.1).

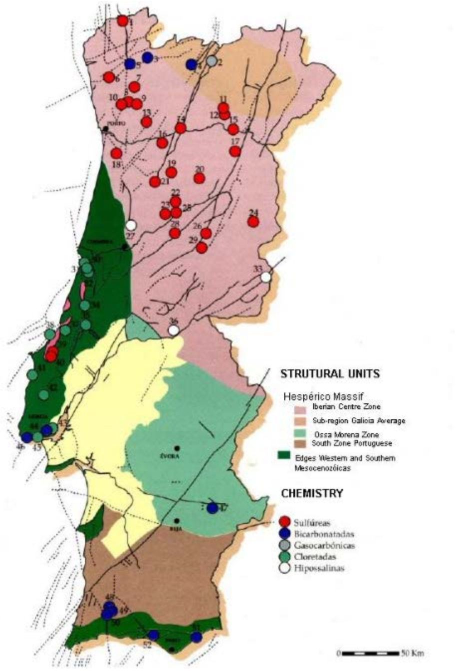

Fig. 1: Thermal centers in Portugal [4].

This work presents the methodology developed for the radiological characterization of selected Portuguese thermal centers based on indoor radon dosimetry. The developed work consisted in the assessment of the effective doses at 16 thermal centers and risk estimation based on the information of radon concentrations. A deterministic approach was used for dose assessment and a probabilistic approach was adopted to estimate the resulting risk by the Monte Carlo method.

MATERIALS AND METHODS

Occupational Exposure in Workplaces

ICRP defines the occupational exposure as all exposures incurred at work as a result of situations that can reasonably be regarded as being the responsibility of the operating management [8].

The recent European Directive 2013/59 EURATOM stipulates a limit on effective dose for occupational exposure of 20 mSv in any single year. Below this dose limit, the principle of optimization requires that any radiation exposure should be kept as low as reasonably achievable (ALARA). When the annual dose limit is exceeded, the regulatory body can permit this exposure by considering the individual case and/or imposing work conditions and dose restrictions for the successive years, providing that the annual dose over any five consecutive years, including the years for which the limit has been exceeded, does not exceed 20 mSv.

The U. S. Environmental Protection Agency evaluates the risk due to radiation exposure as the carcinogenic slope factor, representing the lifetime excess total cancer risk per unit intake or exposure. The cancer slope factor represents the slope of the dose-response curve, at very low concentrations, thus quantifying the cancer inducing potential; the unit is the inverse of a dose. The product of the cancer slope factor by the dose received estimates the risk for a member of the critical group. The risk represents the probability of cancer inducing by this particular exposure, in excess relatively to the background risk, known as the incremental lifetime risk [8].

The radiological risk assessment is an estimate of the probability of a fatal cancer over the lifetime of an exposed individual while a radiological dose assessment calculates the amount of radiation energy that might be absorbed by a potentially exposed individual as a result of a specific exposure.

The acceptable risk is generally defined as 10^{-6} for the general public and 10^{-5} for workers. This means that an additional one case of cancer is accepted for populations of 1 million or 100 000, respectively. A risk level of 1 in a million, or 1 in one hundred thousand, also implies a likelihood that up to one person out of one million (or 100 000) equally exposed people would contract cancer if exposed continuously (24 h/d) to a specific radiation dose over 70 years (average lifetime). This value is in excess to the normal background number of cancer originated by multiple and indeterminate causes, respectively 200 000 or 20 000 [8], [9].

The exposure scenario adopted in this study considers both internal and external exposure for estimating the dose and the associated risk for workers involved in water treatments therapeutics at the selected Portuguese thermal centres. The critical group is represented by an average adult worker assuming an exposure during an 8-hour work day, 5 days per week, 49 weeks/year for an average exposure period of 20 years as most of the employees (79 %) work at the same thermal centre for the last 20 years. A worst-case scenario was considered for exposure frequency assuming that workers have not job rotation.

Radon inhalation as well as the inhalation of dust-borne long-lived alpha emitters, contribute significantly to the effective dose and in this way, the exposure may occur mainly by: i) inhalation of radon and decay products, ii) inhalation of dust-borne long-lived alpha emitters and iii) external radiation. However, in this study only the first and the last exposure pathway were considered due to the lack of accurate data related to long-lived alpha emitter’s attached to dust particles [8].

Design of the Survey

For the purpose of this study 16 thermal centres were selected (out of 38 active) located mostly in the northern part of Portugal. The selection was based on local geology and hydrogeology as well as on the acceptability of the thermal centre to participate in the study [10]. In this region, the geology settings comprises predominantly granite and other plutonic rocks and therefore higher radon concentrations are to be expected for natural reasons.

The presence of several joints and faults are closely related to the occurrence of thermal springs [7], [11] and this was considered as the main natural source of radon for the occupational environment as radon dissolved in water may enter into the indoor air when water is used for treatments.

The concentration of radon was measured at different workplaces of each one of the selected thermal centers. The chosen locations included treatment rooms, pools and some access spaces where workers remain during treatment sessions [4]. The assessment was carried out between November 2013 and September 2015, during two different periods: spring/summer and autumn/winter. A gamma dose rate assessment was also performed at the same locations with measurements for continuous periods between 25 and 45 days. Measurements were taken under normal activities and operating conditions [4], [7].

Radon concentration was also measured in the natural mineral water used for therapeutic purposes in the selected thermal centers. The results can be found in [10], [12].

A worker from each thermal center was selected to be monitored for radon concentration at his own dwelling in order to consider the contribution from outside of the workplaces. The selection took into account mainly the length of service at the present task and the proximity of the thermal centre to the dwelling. In this case, the source of indoor radon was considered to be subsoil and eventually construction materials as there is no usage of thermal water. The indoor radon concentration measurements at workers’ dwellings were conducted for periods of 42 days [4].

Several information concerning both thermal centres and dwellings were collected as well as personal data referring to the selected worker through 3 structured questionnaires and an observation checklist. The questionnaires explores: i) specific characteristics such as age, gender, medical history (illness and declared pathologies) and life style; ii) a characterization of the professional tasks in order to identify the workplace from each centre where the exposure will theoretically be higher, its duration and frequency, and ii) location, type of construction, ventilation rate, number of rooms and usage on both selected thermal centres and workers' dwellings [4], [12].

For dose assessment the adopted methodology follows the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources where its stated that in cases where individual monitoring is inappropriate, inadequate or not feasible, the occupational exposure of the worker shall be assessed on the basis of the results of monitoring of the workplace and information on the locations and durations of exposure of the workers [11], [13].

Radon Measurements and Gamma Dose Rates Analysis

Radon concentration measurements were performed using CR-39 nuclear track detectors enclosed in small cylindrical diffusion chambers (5-cm height, 3-cm diameter) for periods between 25 and 45 days. This detector comprises a small piece of polycarbonate, highly sensitive to ionizing particle tracks such as the alpha particles. Quality assurance measures were followed to minimize further exposures of the detectors before and after usage.

The CR-39 detectors were placed in each room at approximately 2 meters from the floor. After a period of exposure in average of 45 days, the detectors were removed and stored individually in sealed containers to prevent any contamination from others sources during transport to the laboratory [4]. The analysis was performed in the Natural Radioactivity Laboratory in the Department of Earth Sciences of the University of Coimbra.

For the analysis and processing of alpha particles traces at surface, the detectors were etched in 25 % NaOH solution at 90 °C for 270 min. The number of tracks in a 1-cm^2 area on each film was counted by a microscope automatic reader. The background track density was subtracted and related to radon concentration level using a calibration factor obtained by exposure of detectors of the same batch in a certified calibration chamber [4], [14].

The quality of the results were guaranteed by a traceability system set up both for sampling and measuring. In addition, this laboratory takes part regularly in inter-comparison exercises with other laboratories in order to estimate the statistical uncertainty (analytical error less than 10 % of the obtained value). The detection limit using the described procedure is of 5 Bq/m^3 [4], [14].

The indoor radon concentrations were compared with the recommended WHO IAQ guidelines, with the EU reference level of dose exposure in existing exposure situations (Directive 2013/59 EURATOM) and the national protection threshold.

Indoor gamma dose rates were hourly registered with a Geiger counter (Gamma Scout) for a time period between 25 and 45 days.

Effective dose assessment

The inhalation effective dose was calculated in both environments (occupational and residential) as a comparison baseline for the exposure at workplace and dwellings. A deterministic methodology was adopted where all inputs parameters were defined as a single fixed value [8].

The annual effective dose due to radon inhalation (H_{int}, mSv/y) was calculated from the measured value of indoor radon concentration (CRn, Bq/m^3), using the expression given by UNSCEAR [1] (Eq. 1):

$$H_{int} = C_{Rn} \, x \, F \, x \, E \, x \left(DCF\right) \qquad \text{(Eq. 1)}$$

where H_{int} is the annual effective dose (mSv/y), $C_{Rn,}$ (Bq/m^3) is the average radon concentration measured at 2 m from the ground, F is the equilibrium factor between radon and its progeny, E (hours/year) is the occupancy factor and DCF (mSv/y per Bq/m^3) is the dose coefficient factor (effective dose received by adults per unit of ^{222}Rn activity per unit of air volume).

The equilibrium factor for radon decay products (F) is a measure of the degree of radioactive equilibrium between radon and its short-lived radioactive decay products representing the fraction of potential alpha decay energy of the short-lived radon decay products, compared to secular equilibrium. Due to the high variably of radon equilibrium factor, it is common to find a wide range of values for this factor in literature: 0.4 [13], [15], [16]; 0.51 [17]; 0.54 [18] and 0.6 [19], [20].

Radon progeny has a very important contribution to internal exposure (alpha-radiation) and should also be measured beside radon concentration alone. Nevertheless, most of the radon progeny data results from the extrapolation of the radon concentration through the use of equilibrium factors not generally known [1] and the main reason for this is relies on the complexity in determining radon equilibrium factor. An appropriate determination would require long-term measurements in order to account for seasonal and diurnal variations according to meteorological conditions [8].

For the exposure frequency parameter (E) it was considered 1960 hours per year, resulting from the exposure of 8-hour work day, 5 days per week, 49 weeks/year [8]. For the dwellings' exposure, and considering the same individual worker, it was assumed that the rest of time was spent at home (indoors).

The dose coefficient factor (DCF) also known as the radon equilibrium equivalent concentration (EEC) represents the conversion of potential alpha energy exposure (Bq h/m^3) to effective dose equivalent (nSv). The EEC is the concentration of radon that, in equilibrium with each one of the daughters, would have the same potential alpha-energy per unit volume as the actual mixture [8]. A value of 9 nSv/h per Bq/m^3 (9 x 10^{-6} mSv/y per Bq/m^3) was used, according to the recommendations of the United Nations Scientific Committee on the Effects of Atomic Radiations [21].

The external dose (H_{Ext}, mSv/y) is calculated with the measured gamma dose rates (D_γ, mSv/d) and the previous exposure parameters (Eq. 2). The total effective dose is obtained by combining the internal and external doses for comparison with dose limits.

$$H_{Ext} = D_\gamma \times E \qquad \text{(Eq. 2)}$$

Risk Assessment

A probabilistic risk-based approach was used to assess the annual risk incurred to the workers both by internal exposure due to radon inhalation and due to external exposure to gamma radiation.

The annual risk resulting from radon inhalation (R_{Rn}) was calculated by combining the average indoor radon measurements (C_{Rn}, Bq/m^3) with the individual inhalation rate at the exposure location (BR, m^3/d), the exposure frequency (E, d/year) and the radon cancer slope factor (CSF, Risk/Bq) as given by Eq. 3. A single input fixed value of 4.86×10^{-10} (Risk/Bq) was adopted for this parameter [9]. Probabilistic distributions were used for the others input parameters [8].

$$R_{Rn} = C_{Rn} \times F \times BR \times E \times (CSF) \qquad \text{(Eq. 3)}$$

In general, indoor radon data can be approximately described by a log-normal distribution [22] as can be expected from the central limit theorem from a variable which is the product of many independent random factors. In most cases, the interest is focussed on the high-concentration side of the distribution. In this study it was considered that the obtained values for indoor radon follow the log-normal trend.

For the inhalation rate (BR), a log-normal distribution of daily inhalation rate, normalized to the average body weight, was adopted with a mean and standard deviation designated by the International Commission on Radiological Protection [23] of 16.45 and 4.69 m^3/d, respectively [8] [24].

A triangular distribution was adopted for the indoor exposure frequency with 180 d/year (minimum), 307 d/year (maximum), and 245 d/year (most probable) as the input parameters of the distribution [25]. The likeliest value was establish considering that a worker works 2 weekends per year, takes 2 weeks' vacation and 2 weeks' sick leave. The minimum value was establish by assuming that the worker is part-time and only at the site approximately 60 % of the time. The maximum value was based on a person taking 2 weeks' vacation, 2 week's sick leave and working all but 15 weekends per year [25]. For the residential scenario it was assumed that the same individual spends the rest of the time at home: 58 d/year (minimum), 209 d/year (maximum), and 120 d/year (most probable).

The long-term variation of outdoor radon equilibrium factor adjust to a log-normal distribution [26]. A value of 0.51 ± 0.12, for the mean and the standard deviation, were used in this simulation as input parameters of the log-normal distribution [8].

The annual risk induced by external exposure to gamma radiation (R_{Ext}) was obtained, from the calculated values for the external gamma dose exposure (H_{Ext}), with Eq. 4:

$$R_{Ext} = H_{Ext} \times (DRC) \qquad \text{(Eq. 4)}$$

where R_{Ext} is the cancer risk for external exposure due to gamma dose (the health risk from a given radiation dose) and (DRC) is the Dose-to-Risk Conversion Factor for Cancer Mortality (fatal cancer risk per Sievert). For stochastic effects, ICRP 60 [27] uses values of 0.04 Sv^{-1} for workers and 0.05 Sv^{-1} for the public.

The described probabilistic distributions were used in the risk calculation approach, both for internal exposure and external exposure. The Monte Carlo method was applied to generate the distribution for the input parameters as well as an output distribution of the resulting risk, developed in Matlab® code.

RESULTS AND DISCUSSION

Radon concentration

The results of indoor radon concentrations measured in the selected thermal centres (TC) and in the selected workers' dwelling (DW) are presented in Table I (mean - µ, SD - standard deviation, range - Rg, median - Md). For dwellings, a single measurement was carried out and it was not possible to take measurements within the dwellings of workers from the TC2 and TC12 [4].

TABLE I: Summary statistics of radon concentration (TC and DW)

TC/DW Nº	**n**	**µ (TC) (Bq/m³)**	**SD (TC) (Bq/m³)**	**Rg (TC) (Bq/m³)**	**Md (TC) (Bq/m³)**	**µ (DW) (Bq/m³)**
1	6	1189	1232	238-3479	562	68
2	3	569	116	422-707	577	n.a.
3	9	364	210	152-724	267	254
4	5	428	70	274-502	445	1322
5	2	3124	1212	1912-4335	3124	312
6	6	1256	430	878-2181	1168	1877
7	5	1047	564	366-1681	1148	168
8	2	354	7	347-361	354	642
9	2	262	107	143-376	265	105
10	3	233	90	121-406	217	714
11	5	480	255	209-1079	377	4051
12	3	1122	403	813-1692	862	n.a.
13	4	187	136	73-498	124	257
14	6	2090	553	1130-2873	2298	605
15	3	166	46	93-235	161	111
16	5	288	100	172-467	253	508
Total/Mean	**69**	**721**	**809**	**73-4335**	**403**	**10.38**

n - number of locations within the same thermal spa; *n.a.* - not available

For thermal centres, radon mean levels are widely distributed from 166 to 3124 Bq/m^3; the median ranged from 124 to 3124 Bq/m^3. The standard deviation is relatively high which may be explained by the high variability of the sampling locations within the thermal centre in addition to the geographical location of each thermal centre.

Ninety-seven per cent of thermal centres reported indoor radon levels higher than 100 Bq/m^3, the WHO reference limit to minimize health hazards from indoor radon exposure [12] (the mean values are all above 100 Bq/m^3). In 69 % of the thermal centres the mean radon concentrations exceeded the EU maximum reference level of 300 Bq/m^3. In what concerns to the national action level 56 % of the thermal centres presented mean indoor radon concentration higher than 400 Bq/m^3 and 38 % exhibited mean values higher than 100 Bq/m^3.

Also, radon concentration at different points in the thermal centres presented very high variability. Heterogeneous distribution of concentrations were observed at all treatments rooms of the thermal centres. In general, the highest values were registered in the inhale-therapy rooms (ORL) followed by the Vichy showers and pools. High radon levels (1145-1692 Bq/m^3) were also registered in technical rooms where thermal waters are not used [11], [12], [29].

For workers' dwellings, radon concentration levels are of the same order of magnitude as for the thermal centres, ranging between 68 and 4051 Bq/m^3. In 93 % of the dwellings, radon concentration exceed the WHO reference value of 100 Bq/m^3, 73 % exceed the EU maximum reference level of 300 Bq/m^3 and 50 % exceed the national action level of 400 Bq/m^3.

Dose assessment

For the considered scenario (average radon concentration for each thermal center and no job rotation) and using a deterministic approach, the mean annual dose received by the workers due to radon inhalation ranged between 1 mSv/y and 28 mSv/y [7].

The results obtained for the gamma dose rate measurements, carried out simultaneously with the measurements of indoor radon concentration within the thermal centres, are presented in table II.

TABLE II: Summary statistics of gamma dose rate measurements within the studied thermal centers (TC)

TC Nº	**µ (mSv/y)**	**SD (mSv/y)**	**Rg (mSv/y)**	**Md (mSv/y)**
1	2.73	2.40	1.30-4.95	1.95
2	3.05	0.32	2.73-3.59	2.81
3	4.24	0.92	2.24-7.98	3.54
4	2.54	1.73	2.18-2.88	2.56
5	2.79	0.18	1.91-6.50	2.40
6	4.29	0.22	1.29-4.29	4.29
7	3.47	0.40	3.24-3.71	3.46
8	2.51	2.12	1.16-3.88	3.61
9	3.76	0.35	3.65-3.87	3.76
10	3.67	0.46	2.51-4.83	3.67
11	2.48	0.08	2.04-2.83	2.58
12	3.56	0.20	2.89-5.52	3.44
13	2.57	1.80	2.54-2.60	2.57
14	3.00	0.08	2.91-3.08	3.00
15	3.67	2.40	2.45-9.08	2.69
16	3.38	0.32	2.50-5.02	2.61
Total	**3.25**	**1.22**	**1.30-9.08**	**2.82**

The total effective dose was obtained by combining the doses resulting from internal and external exposures; the results ranged between 4 and 31 mSv/y. The results show that the external exposure resulting from the gamma dose rate is negligible when comparing with the inhalation dose. Moreover, the inhalation dose is underestimated as dust inhalation was not included in the dose calculation.

The recent Directive 2013/59/EURATOM, defines the basic standards for the protection of the workers and the general public against the dangers arising from the ionizing radiation.

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 which are defined by: 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 [7], [9].

For workers' dwellings the annual dose ranged between 1 and 54 mSv/y which is above the range of what was observed for thermal centres and above the limit for the general public of 1 mSv/y.

Risk Assessment

Probabilistic distributions of the input parameters used in risk calculations (radon inhalation risk and external gamma radiation risk) are presented in Fig. 2.

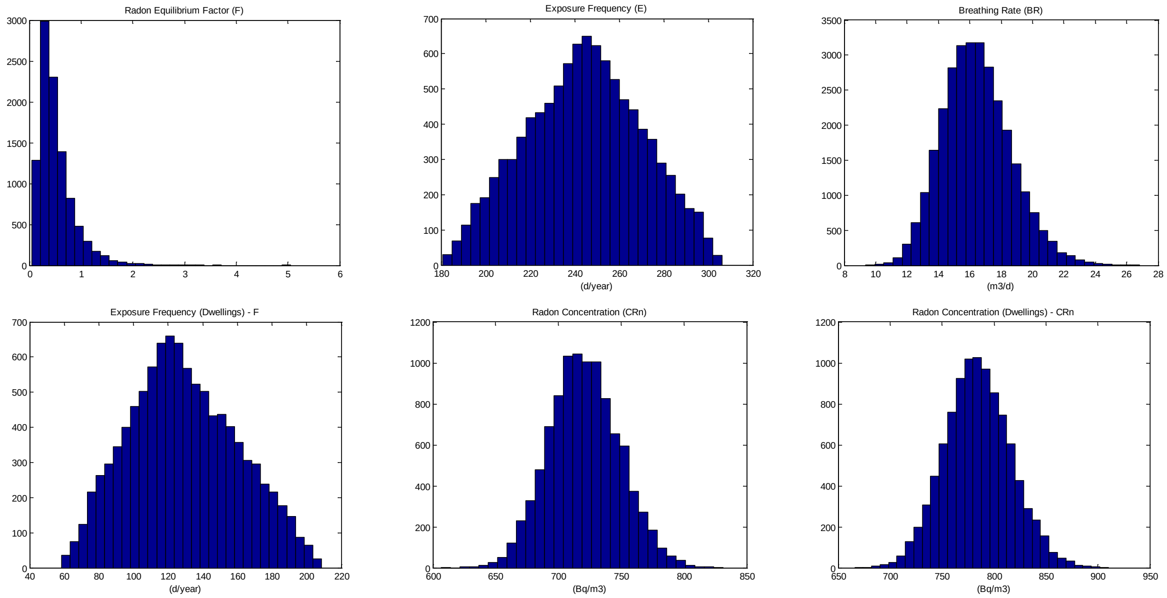

Fig. 2: Probabilistic distributions of the input parameters for risk calculations.

The distribution of radon concentration was generated for each one of the thermal centres with the respective median and standard deviation. As all the distributions have the same shape, only the distribution representing the mean of all thermal centres is presented here. The same approach was adopted for the workers' dwellings, correcting the exposure time/frequency.

The probabilistic distributions of the calculated risk are presented in Fig. 3 for workers' internal exposure, in Fig. 4 for external exposure at thermal centres (TC), and in Fig. 5 for internal exposure at worker's dwellings (DW).

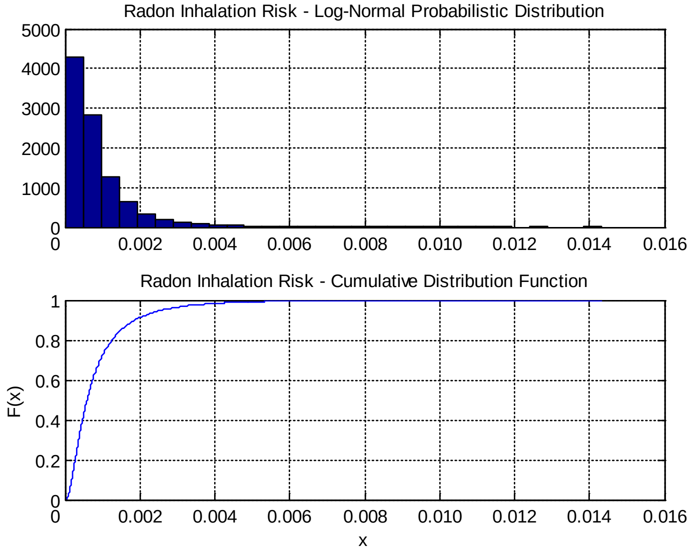

Fig. 3: Radon inhalation risk: log-normal probabilistic distribution and cumulative function (TC).

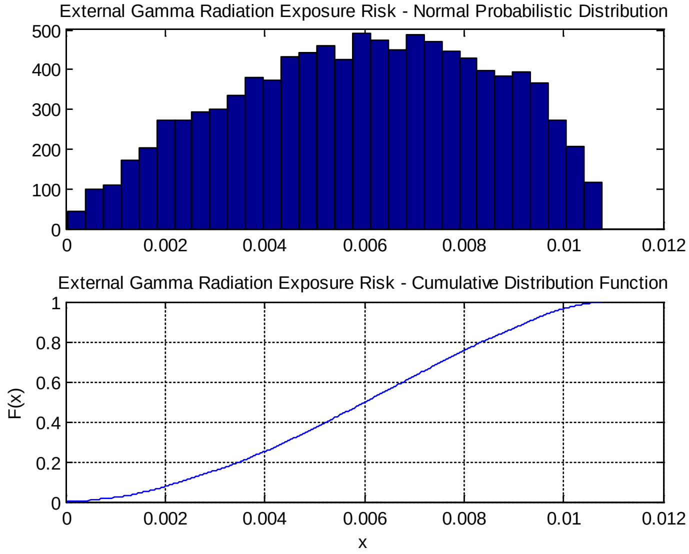

Fig. 4: External gamma radiation exposure risk – normal probabilistic distribution and cumulative probability functions (TC).

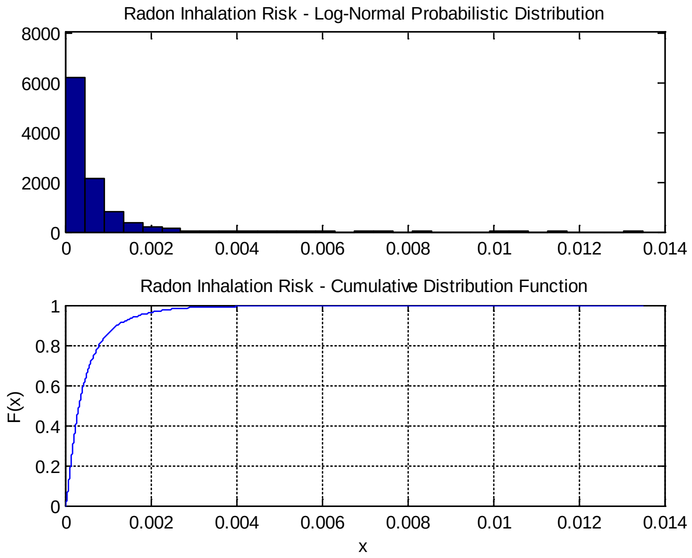

Fig. 5: Radon inhalation risk: log-normal probabilistic distribution and cumulative function (DW).

The results from the probabilistic approach, concerning the adopted exposure scenario, are summarized in Table III. Two descriptors are presented for the characterization of the risk: central tendency (mean - µ and median - Md) and high-end estimate (95th percentile – 95th) of individual risk.

TABLE III: Annual risk from radon inhalation and external exposure (TC and DW)

Nº TC/DW	Annual risk - radon inhalation (TC)			Annual risk - external exposure (TC)			Annual risk - radon inhalation (DW)		
	µ	Md	95th	µ	Md	95th	µ	Md	95th
1	$1x10^{-3}$	$1x10^{-3}$	$4x10^{-3}$	$5x10^{-6}$	$5x10^{-6}$	$8x10^{-6}$	$5x10^{-5}$	$3x10^{-5}$	$1x10^{-4}$
2	$7x10^{-4}$	$5x10^{-4}$	$2x10^{-3}$	$6x10^{-6}$	$6x10^{-6}$	$9x10^{-6}$	*n.a.*	*n.a.*	*n.a.*
3	$4x10^{-4}$	$3x10^{-4}$	$1x10^{-3}$	$8x10^{-6}$	$8x10^{-6}$	$1x10^{-5}$	$2x10^{-4}$	$1x10^{-4}$	$5x10^{-4}$
4	$5x10^{-4}$	$3x10^{-4}$	$1x10^{-3}$	$5x10^{-6}$	$5x10^{-6}$	$8x10^{-6}$	$9x10^{-4}$	$6x10^{-4}$	$3x10^{-3}$
5	$4x10^{-3}$	$2x10^{-3}$	$1x10^{-2}$	$5x10^{-6}$	$5x10^{-6}$	$8x10^{-6}$	$2x10^{-4}$	$1x10^{-4}$	$7x10^{-4}$
6	$1x10^{-3}$	$1x10^{-3}$	$4x10^{-3}$	$8x10^{-6}$	$8x10^{-6}$	$1x10^{-5}$	$1x10^{-3}$	$8x10^{-4}$	$4x10^{-3}$
7	$1x10^{-3}$	$9x10^{-4}$	$3x10^{-3}$	$6x10^{-6}$	$7x10^{-6}$	$1x10^{-5}$	$1x10^{-4}$	$7x10^{-6}$	$4x10^{-4}$
8	$4x10^{-4}$	$3x10^{-4}$	$1x10^{-3}$	$5x10^{-6}$	$5x10^{-6}$	$8x10^{-6}$	$4x10^{-4}$	$3x10^{-4}$	$1x10^{-3}$
9	$3x10^{-4}$	$2x10^{-4}$	$9x10^{-4}$	$7x10^{-6}$	$7x10^{-6}$	$1x10^{-5}$	$7x10^{-5}$	$4x10^{-5}$	$2x10^{-4}$
10	$3x10^{-4}$	$2x10^{-4}$	$8x10^{-4}$	$7x10^{-6}$	$7x10^{-6}$	$1x10^{-5}$	$5x10^{-4}$	$3x10^{-4}$	$1x10^{-3}$
11	$6x10^{-4}$	$4x10^{-4}$	$2x10^{-3}$	$5x10^{-6}$	$5x10^{-6}$	$8x10^{-6}$	$3x10^{-3}$	$2x10^{-3}$	$8x10^{-3}$
12	$1x10^{-3}$	$9x10^{-4}$	$4x10^{-3}$	$6x10^{-6}$	$7x10^{-6}$	$1x10^{-5}$	$2x10^{-4}$	$1x10^{-4}$	$6x10^{-4}$
13	$2x10^{-4}$	$1x10^{-4}$	$7x10^{-4}$	$5x10^{-6}$	$5x10^{-6}$	$8x10^{-6}$	*n.a.*	*n.a.*	*n.a.*
14	$2x10^{-3}$	$2x10^{-3}$	$7x10^{-3}$	$5x10^{-6}$	$6x10^{-6}$	$9x10^{-6}$	$4x10^{-4}$	$3x10^{-4}$	$1x10^{-3}$
15	$2x10^{-4}$	$1x10^{-4}$	$6x10^{-4}$	$7x10^{-6}$	$7x10^{-6}$	$1x10^{-5}$	$8x10^{-5}$	$5x10^{-5}$	$2x10^{-4}$
16	$3x10^{-4}$	$2x10^{-4}$	$1x10^{-3}$	$6x10^{-6}$	$6x10^{-6}$	$1x10^{-5}$	$3x10^{-4}$	$2x10^{-4}$	$1x10^{-3}$

In general, the results indicate that radon inhalation poses a higher risk than external radiation and in this way, inhalation is considered as the main exposure pathway.

The highest inhalation risk correspond to TC5 and TC14: a 50th percentile of 0.0025 ($\cong 2x10^{-3}$) and 0.0017 ($\cong 2x10^{-3}$), respectively; a 95th percentile of 0.0108 ($\cong 1x10^{-2}$) and 0.0071 ($\cong 7x10^{-3}$), respectively.

For external exposure, all thermal centres present a risk of magnitude 10^{-6} with higher values corresponding to TC3, TC6, TC9, TC10 and TC15.

Considering the inhalation risk at worker's dwelling, higher values were obtained for DW6 and DW11: a 50th percentile of 0.000783 ($\cong 8x10^{-4}$) and 0.00170 ($\cong 2x10^{-3}$), respectively; a 95th percentile of 0.004 ($\cong 4x10^{-3}$) and 0.008 ($\cong 8x10^{-3}$), respectively.

As expected, higher risk values were obtained for radon inhalation (TC) and this should be even more pertinent if dust inhalation (radon progeny) was considered.

Gamma dose rates were not assessed at workers' dwellings and although the values obtained for the risk due to external gamma exposure at thermal centres was within the range [10^{-5}, 10^{-6}] this should also be assessed at dwellings in order to achieve a complete characterization of the exposure.

The estimative of intake by inhalation and external exposure over the period of the estimated exposure (20 years for thermal centres and 30 years for dwellings) is calculated by combining this exposure duration with the annual risk presented in Table III. The total incremental lifetime cancer risk is then estimated as the sum of the risks in all exposure pathways.

CONCLUSIONS

The results from this study showed that several reference levels (indoor radon levels, effective dose) were exceed both at occupational and residential environments. In addition, there are several cases when the reference level of "an existing exposure situation" of 6 mSv/y is also exceed, and in these cases, according to the EU Directive 2013/59/EURATOM, the exposure should be classified as "a planned exposure situation" and actions should be taken. The results at worker's dwellings are also worrisome being sometimes higher than in the occupational environments.

According to indoor radon concentration there are 9 thermal centres (out of 16) and 7 dwellings (out of 14) with values above 400 Bq/m^3. Approximately 80% of the total measurements of indoor radon concentration exceeded the EU reference level of 300 Bq/m^3 (Directive 2013/59/EURATOM).

For the effective dose, there are 11 thermal centres with values above 6 mSv/y and for all workers' dwellings, the effective dose (inhalation) exceeded the limit of 1 mSv/y for the general public. From the results obtained in Table III, is possible to identify the situations with higher risk where an urgent action must be addressed. Such places must be subjected to special and more detailed investigation, taking into account the whole mobility of radon and progeny in the ambient air. The probabilistic estimates of risk should be presented as a supplement of the deterministic approach as a probabilistic approach may be used to provide quantitative estimates of the uncertainties in the risk assessment.

REFERENCES

1. UNSCEAR 2000, "*Sources and effect of ionizing radiation*". United Nation Scientific Committee of the Effect Atomic, Radiation Report on The General Assembly, United Nation, New York (2000).
2. ICRP 1994, *"Protection against 222Rn at home and at work"*, ICRP Publication 65, Ann ICRP, 23 (1994).
3. WHO, *Handbook on indoor radon: a public health perspective"*, Ed. H. Zeeb & F. Shannoun, ISBN 978 92 4 154767 3 (2009).
4. Silva, A., Dinis, M.L., Pereira, A.J.S.C., *"Assessment of indoor radon levels in Portuguese thermal spas"*, Radioprotection, EDP Sciences, doi: http://dx.doi.org/10.1051/radiopro/2016077 (2016).
5. ICRP, *"Lung Cancer Risk from Radon and Progeny and Statement on radon"*, ICRP Publication 115 Ann, ICRP 40(1) (2010).
6. Silva, A.S., Dinis M.L., *"The presence of radon in thermal spas and their occupational implications – a review"*, Book chapter in: Occupational Safety and Hygiene III, Eds. P. Arezes, J. S. Baptista, M. Barroso, P. Carneiro, P. Cordeiro, N. Costa, R. Melo, A. S. Miguel, G. Perestrelo, pp. 353-355, ISBN 978-1-138-02765-7, London: Taylor & Francis (2015).
7. Silva, A.S., Dinis, M.L., *"Measurements of indoor radon and total gamma dose rate in Portuguese thermal spas"*, Book chapter in: Occupational Safety and Hygiene IV, Eds. P. Arezes, J. S. Baptista, M. Barroso, P. Carneiro, P. Cordeiro, N. Costa, R. Melo, A. S. Miguel, G. Perestrelo, pp. 485-489, ISBN 9781138029422, London: Taylor & Francis, DOI: 10.1201/b21172-93 (2016).
8. Dinis, M.L., Fiúza A., *"Occupational exposure during remediation works at a uranium tailings pile"*, Journal of Environmental Radioactivity, Special Issue on Radioactive Material, Vol. 119, Pages 63–69 (2012), http://dx.doi.org/10.1016/j.jenvrad.2012.08.009.
9. EPA, *"Health Effects Assessment Summary Tables, Radionuclide Carcinogenicity Slope Factors: HEAST"* (1997).
10. Silva, A.S., Dinis, M.L., Pereira, A.J.S.C., Fiúza, A., *"Radon levels in Portuguese thermal spas"*, RAD Journal, Radiation and Applications in Physics, Chemistry, Biology, Medical Sciences, Engineering and Environmental Sciences, pp. 76-80 (2016), doi: 10.21175/RadJ.2016.01.14.
11. Silva, A.S., Dinis M.L., Fiúza, A., *"Research on Occupational Exposure to Radon in Portuguese Thermal Spas"*, Book chapter in: Occupational Safety and Hygiene II, ed. P. Arezes, J. S. Baptista, M. Barroso, P. Carneiro, P. Cordeiro, N. Costa, R. Melo, A. S. Miguel, G. Perestrelo, pp. 323 -328. ISBN: 978-1-138-00144-2, London: Taylor & Francis (2014).
12. Silva, A.S., *"Occupational Exposure to Radon in Thermal Establishments"*, Doctoral dissertation on Occupational Safety and Health, Faculty of Engineering, University of Porto (2016).
13. IAEA, *"Radiation Protection against Radon in Workplaces other than Mines"*, IAEA Safety Reports Series, No.33 (2003).
14. Madureira, J., Paciência, I., Rufo, J., Moreira, A., de Oliveira Fernandes, E., Pereira A., *"Radon in indoor air of primary schools: determinant factors, their variability and effective dose"*, Environ Geochem Health 38, 523-533 (2016).

15. ARPANSA, *“Methods for Monitoring, Assessing and Recording Occupational Radiation Doses in Mining and Mineral Processing”*, Radiation Protection Series Publication Nº 9.1 (2010).
16. MOSHAB, *“Mines Occupational Safety and Health Advisory Board”*, Approved Procedure For Dose Assessment Guideline RSG05, ZMR001DU/R (1997).
17. Koo, V.S.Y., Yip, C.W.Y., Ho, J.P.Y., Nikezic, D. Yu, K.N., *“Experimental study of track density distribution on LR115 detector and deposition fraction of ^{218}Po in diffusion chamber”*, Nuclear Instruments and Methods in Physics Research Section A, Vol. 491, Issue 3, pp. 470-473 (2001).
18. Kojima, H., *“The equilibrium factor between radon and its daughters in the lower atmosphere”*, Environment International, Vol. 22, Suppl. 1, pp. S187-S192, 1996, PI1 SOI60-4120(96)00107-9 (1996).
19. Chen, J., Marro, L., *“Assessment of radon equilibrium factor from distribution parameters of simultaneous radon and radon progeny measurements”*, Radiat Environ Biophys (2011) 50:597–601, doi: 10.1007/s00411-011-0375-8 (2011).
20. Lewis, R.K. and Houle, P.N., *“A living radon reference manual”*, Pennsylvania Department of Environmental Protection, Bureau of Radiation Protection, Radon Division, University Educational Services, Inc. (2009).
21. Chambers, D.B., Lowe, L.M., Stager R.H., *“Long Term Population dose due to Radon from Uranium Mill Tailings”*, The Uranium Institute, Twenty Third Annual International Symposium (1998).
22. UNSCEAR, *“Report to the General Assembly”*, with Scientific Annexes, Annex E, Sources-to-effects assessment for radon in homes and workplaces, ISBN 978-92-1-142274-0, pp. 309 (2006).
23. Cinelli, G., Tondeur F., *“Log-normality of indoor radon data in the Walloon region of Belgium”*, Journal of Environmental Radioactivity, 143, 100-109 (2015).
24. Beals, J.A.J., Funk, L.M., Fountain, R., Sedman, R., *“Quantifying the Distribution of Inhalation Exposure in Human Populations: Distribution of Minute Volumes in Adults and Children*”, Environmental Health Perspectives, Vol. 104, Nº 9, (1996).
25. EPA, *“Exposure Factors Handbook: 2011 Edition”,* EPA/600/R-090/052F, 09/2011 (2016).
26. EPA, *“Risk-based PRGs for Radioactive Contaminants of Risk Assessment Guidance for Superfund (RAGS)”*, Chapter 4, Part B (RAGS Part B), December 1991, OSWER Publication EPA/540/R-92/003 (1991).
27. Holtz, H., Winkler, R., *“Long-term variations of outdoor radon equilibrium equivalent concentration*”, Radiation and Environmental Biophysics, Vol. 33, Number 4, 381-392, doi: 10.1007/BF01210459 (1994).
28. ICRP, *“1990 Recommendations of the International Commission on Radiological Protection”*, ICRP Publication 60, Ann. ICRP 21 (1-3) (1991).
29. Silva, A.S., Dinis, M.L., *“Evaluation of the Concentration of Radon in the Natural Mineral water of Portuguese Thermal Spas”*, Book Occupational Safety and Hygiene III, Eds. P. Arezes, J.S. Baptista, M. Barroso, P. Carneiro, P. Cordeiro, N. Costa, R. Melo, A.S. Miguel, G. Perestrelo, pp. 353-355, ISBN 978-1-138-02765-7, London: Taylor & Francis (2015).