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Radon Levels in Nurseries and Primary Schools in *Bragança* district – Preliminary Assessment

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Abstract

Lung cancer has been associated with radon concentration even at low levels as those found in dwellings. This study aimed to: i) determine radon diurnal variations in three nurseries and one primary school at *Bragança* district (North of Portugal); and ii) compare radon concentrations with legislated standards and assess the legislated procedures. Radon was measured in three nurseries and a primary school at a rural area with non-granite soil. Measurements were performed continuously to examine differences between occupation and non-occupation periods. Indoor temperature and relative humidity were also measured continuously. A great variability was found in radon concentrations between microenvironments examined. Radon concentrations surpassed several fold the recommended guidelines and thresholds and excessive levels of health concern were sporadically found (361.5-753.5 Bq m^{-3}). Thus it is of importance to perform a national campaign on radon measurements and to reduce exposure.

Keywords

Radon concentration, nurseries, radon legislation

Introduction

Epidemiological studies in Europe, North America and Asia provided strong evidence of an association between indoor radon exposure and lung cancer in the general population, even at the relatively low radon levels commonly found in residential buildings (WHO, 2009; Tracy et al., 2006; Tong et al, 2012). Radon is considered the leading cause of lung cancer among non-smokers and after tobacco is the second greatest cause of lung cancer in the general population. There is no known threshold concentration below which radon exposure presents no risk; the dose-response relation is linear (USEPA, 2010; WHO, 2009; Zielinski et al., 2006). Current estimates of the proportion of lung cancers attributable to radon range from 3 to 14%, depending upon the average radon concentration in the country concerned and calculation methods (WHO, 2009). Considering the latest scientific data, the World Health Organization (WHO) proposed a reference level of 100 Bq/ m^3 for indoor radon to minimize health hazards. However, if this level can not be reached under prevailing country-specific conditions, the reference level is set at 300 Bq/ m^3. The United States Environmental Protection Agency (USEPA) recommended not exceeding 150 Bq/ m^3. The European Union (EU) proposed annual reference levels of 400 Bq/ m^3 for existing buildings and 200 Bq/m^3 for new ones (EU, 90/143/Euratom). Recently, EU proposed that the reference levels for the annual average activity concentration in air should not exceed 300 Bq/ m^3 (Council Directive 2013/59/Euratom). Some countries defined policies regarding radon exposure in homes: i) Luxemburg sets it to 150 Bq/ m^3; ii) Ireland, UK, Spain and Sweden establish the action level at 200 Bq/ m^3 (in Sweden remediation measures are compulsory when this level is exceeded in dwellings); and iii) in Germany, Belgium, Finland and Austria the action level depends upon conditions of the dwelling, being 400 Bq/m^{-3} for the existing ones and 200 Bq/ m^3 for new ones, as recommended by the EU (Antão, 2014; Iglesias and Taboada, 2014). Portuguese legislation defines a limit of 400 Bq/ m^3 and mandates measurements in the districts of *Braga*, *Vila Real*, *Porto*, *Guarda*, *Viseu* and *Castelo Branco*, areas with granite soils, which may present higher risk (Portaria nº 353-A/2013).

Several studies reported radon levels in granite regions in countries such as Spain, Romania, Iran and Norway (Sainz et al., 2009). Despite this, indoor radon problems may be generated by other types of rocks such as carbonaceous black shales, glauconite-bearing sandstones, certain kinds of fluvial sandstones and fluvial sediments, phosphorites, chalk, karst-producing carbonate rocks, bauxite, lignite, graphitic schist and slate, silica-rich volcanic rocks, and certain kinds of contact-metamorphic rocks (Gundersen et al., 1992). In addition, building materials may also exert influence on radon concentrations. Further, studies in Norwegian dwellings revealed that the entry of radon from the building ground is the predominant source of indoor radon (Sundal, 2003).

Studies in Spain demonstrated concentrations as high as 800 Bq/ m^3 in *Santiago de Compostela* (university buildings), several times exceeding 200 Bq/ m^3 in dwellings of the Galician region and a mean of 43 Bq/ m^3 in the Spanish territory (Iglesias and Taboada, 2014). Studies in Iran noted median concentrations in dwellings between 62 and 126 Bq/ m^3 (Hadad and Mokhtari, 2015). Celebi et al. (2014) reported indoor radon concentrations in Turkey between 1 and 1400 Bq/ m^3 (mean: 81 Bq/ m^3) measured in 7293 dwellings with passive detectors where 1% of dwellings surpassed 400 Bq/ m^3. Ramachandan and Sathish (2011) reported a nationwide radon map for India, where more than 5000 measurements were carried out in 1500 dwellings across the country comprising urban and non-urban locations with passive detectors. Results showed geometric means between 4.6 and 147.3 Bq/ m^3 with an overall geometric mean of 23 Bq/ m^3. Levels of radon were also measured in Central Portugal (*Coimbra*, *Viseu*, *Castelo Branco*, *Guarda*) and found to be variable and several fold higher than 200 Bq/ m^3 (1-3 months measurements with concentrations at ground floor rooms higher than 400 Bq/ m^3) (Antão, 2014). Most studies were performed in dwellings but there were also some conducted in schools. However, there are no apparent studies in nurseries. Kapdan and Altinsoy (2012) reported a study performed with passive radon detectors comparing dwellings and schools in Turkey and concluded that these were similar with a mean around 60 Bq/ m^3. Clouvas et al. (2011) conducted radon measurements in 512 schools in 8 of the 13 regions of Greece. Most of the radon concentrations (86%) were between 60 and 250 Bq/ m^3 with a most probable value of 135 Bq /m^3. The arithmetic and geometric means of radon concentration were 149 Bq/ $m^3$and 126 Bq/ m^3, respectively. Kitto (2014) noted radon measurements at 186 schools in New York also with passive samplers. Results showed that the majority of rooms contained radon levels below 148 Bq/ m^3 and less than 10% of all measured rooms exceeded this level.

Most studies were performed using passive samplers and none conducted in nurseries. Thus, this study as part of the INAIRCHILD Project (Sousa et. al., 2012) aimed to: i) determine radon diurnal variations in three nurseries and one primary school at *Bragança* district (North of Portugal); and ii) compare radon concentrations obtained with legislated standards.

Materials and Methods

Measurements were performed in three nurseries (1 with children from 0 to 2 years old – N_RUR_2 and 2 with children from 3 to 5 years old – N_RUR_1 and N_RUR_3) and one primary school – PRIM_RUR_1 - at *Bragança* district, in the North of Portugal in November and December 2013. N_RUR_1 and PRIM_RUR_1 are located in the same building. All sites are considered rural and with low traffic influence. N_RUR_1, PRIM_RUR_1 and N_RUR_2 were built in an area of contact-metamorphic rocks and

N_RUR_3 in an area of silica-rich volcanic rocks (LNEG, 2013) and all were constructed of concrete and/or brick and/or wooden materials. Table 1 shows the main characteristics of each studied microenvironment and sampling periods.

A prior inspection to the studied nurseries and rooms (observations and staff interviews) was developed to capture relevant information on activities, ventilation habits and building characteristics. All studied buildings were a single floor. Only N_RUR_2 had a mechanical ventilation system (HVAC); the others had natural ventilation. During occupation periods, electric/oil heating systems were usually turned on, and windows to outdoor as well as doors to inner corridors were always closed to avoid heat loss. All buildings were completely closed at night and weekends.

Measurements were carried out continuously (logging hourly means), during 2 to 4 days in several rooms of the nurseries and primary school, including weekdays and weekends. Radon levels were measured with Radim 5B monitor (Jiří Plch – SMM, Czech Republic), which measures the α-activity of radon decay products (^{218}Po and ^{214}Po) collected from the detection chamber on the surface of a semiconductor detector by an electric field. The monitor was placed inside each room at the approximate breath height of the children. Indoor temperature (T) and relative humidity (RH) were measured with electrochemical sensors from the Haz-Scanner IEMS Indoor Environmental Monitoring Station (SKC Inc., USA).

The mean radon indoor concentrations were compared with reference standards and guidelines aiming to evaluate exceedances. Comparisons were performed for occupation periods considering international references, namely from USEPA, WHO and EU – all for 1 hr means, and also considering Portuguese legislation (Portaria nº 353-A/2013) - for 8 hr means. The 8 hr means were calculated as moving averages and comparisons were performed with the daily maxima during occupation periods. T and RH were also compared with ASHRAE guideline ranges (ASHRAE, 2007): i) T - 20-23.9 ºC in winter season; and ii) RH - 30-60%.

Results and Discussion

Table 2 summarizes the main statistical parameters (minimum, maximum, mean and median) of the hourly mean for each room of the three nurseries and primary school. As previously observed for other pollutants (Branco et al., 2014), radon daily concentration patterns in two different sampled weekdays in each room were similar, hence a mean weekday profile was calculated for radon using the hourly mean values. The same was made for T and RH. As with weekdays, the weekend profiles for both radon, T and RH were also similar in Saturdays and Sundays, therefore a mean weekend profile was also calculated.

The daily mean profiles of T and RH are shown, respectively, in Figures 1 and 2 for (a) weekdays and (b) weekends. T and RH for baby classroom (0Y) of N_RUR_2 are not shown due to instrument errors. On weekdays an increase in T was noted during occupation periods, while on weekends identical variation could not be observed, although some variation was detected in the daily profiles probably due to air infiltration through loose fitting windows and doors, thus showing importance of outdoors. The classroom with highest T was the 2 year-old classroom of N_RUR_2 (means between 21-25 ºC) while lowest was the lunch room of PRIM_RUR_1 (means between 16-20 ºC).

Concerning RH, on weekdays a minor decrease was noted in the beginning of the occupation period, due to door opening allowing the exchange with the air from corridors and from outdoor, followed by a rise during the occupation period, probably due to the lack of ventilation associated with accumulation related with occupation. An exception to this profile was found for the 2 years old classroom in N_RUR_2 in which RH increased during all the occupation period. On weekends a numerical RH decrease was found in the morning being more accentuated in the 3rd grade classroom of PRIM_RUR_1, most probably, and according to what occurred with T, due to outdoor influence. The highest RH was observed in the 1st grade classroom of PRIM_RUR_1 (means between 52-66%) and lowest in the 2 year-old classroom of N_RUR_2 (means between 27-44%).

Figure 3 (a) and (b) show daily mean profiles of radon concentrations, respectively for weekdays and weekends. Radon concentrations were generally higher on weekends than weekdays, and in some cases in non-occupied periods of the week probably due to the lack of ventilation during those periods, leading to radon accumulation. It was not possible to determine a well-defined daily pattern for radon concentrations, although these seemed to be higher in the early morning. Neves et al. (2009) who measured radon concentrations in a building of the University of Coimbra (Portugal), also reported maximum concentrations occurring more frequently between 9 and 10 a.m. The highest concentrations were found in the 3rd grade classroom of PRIM_RUR_1. It is noteworthy that N_RUR_1 and PRIM_RUR_1 are located at the same building (same floor), same building materials, with similar ventilation profiles; however, different rooms had different concentrations (mean differences reaching 490 Bq/ m^3). This may be attributed to the position of a potential crack or other type of hole in the building foundations. Radon daily variability seemed to be dependent upon the ventilation profiles, although no correlation was found with indoor T and RH. In some classrooms there was a tendency for concentrations to be lower during occupation, namely on RUR_1 nursery and primary school, due to the higher air renovation with the children transit. Kapdan and Altinsoy (2012) also reported lower concentrations related with ventilation from windows and doors being kept open. N_RUR_2 was the site with lower radon concentrations, both on weekdays and weekend, which was likely due to the use of HVAC. This variability enhances the importance of the need for radon surveillance in all occupied rooms.

Table 3 shows the non-compliances and exceedances (%) of hourly mean values according to ASHRAE, for T and RH, and USEPA, WHO, EU and Portuguese legislation for radon. Temperature was several times out of the ASHRAE proposed range (20-23.9ºC) and RH was generally within the ASHRAE proposed range (30-60%). It was common to find lower temperatures than those recommended by ASHRAE, and thus it is tempting to assume that children might have experienced thermal discomfort which may have affected their attention. Nevertheless, for a detailed evaluation of the thermal comfort conditions, international indices based on T and RH, like those of ISO 7730:2006, need to be calculated. Regarding the EU recommendation and Portuguese legislated standard for radon, only the 3rd grade classroom of PRIM_RUR_1 presented exceedances (88 and 100%, respectively) during the occupation periods. Although high concentrations were found in a great number of rooms, those detected in the 3rd grade classroom of the primary school are extremely worrisome as they were particularly high (means between 362-754 Bq/ m^3). Nevertheless, considering USEPA (150 Bq/ m^3) and WHO guidelines (100 Bq/ m^3) almost all classrooms presented exceedances (between 14 and 100%).

Conclusions

Radon concentrations found in the nurseries and primary school exceeded several times the recommended guidelines and thresholds (WHO- 100 Bq/ m^3; Portaria nº353-A/2013- 400 Bq/ m^3) and levels considered to be a health concern were found (362-754 Bq/ m^3) in one of the classrooms. The variability found between classrooms and in some cases between periods of the day puts into question the effectiveness of a discrete, single measurement, as these measurements may mask continuously changing radon concentrations.

Bragança is not a mandatory area for radon measurement, but still displayed exceeded concentrations, which seemed to be related to the type of soil where buildings were constructed (not granite), thus there might be other areas with the same profile. Considering the results obtained, it might be of great importance to perform a national campaign not only in dwellings, but also in school environments, with special attention to nurseries, which will enable estimation of the effective dose and cancer risk assessment due to radon exposure. In addition, measures to reduce exposure are essential, such as the simple implementation of active ventilation which may reduce concentrations from 30 to 70% (WHO, 2009) and consequently reduce children's exposure and cancer risk.

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Table 1. Main characteristics of each studied microenvironment and sampling periods.

Site	Construction Year	Room (children age)	Room acronym	Area (m^2)	Occupation (# children)	Occupation period	Sampling period (week+weekend)
N_RUR_1	2011	CR[1] (3-5Y)	4-6Y	46.9	24	9h-12h, 13h30-18h30	2+2
N_RUR_2	2010	CR (0Y)	0Y	17.1	9	7h45-20h	2+0
		CR (2Y)	2Y	20.6	15	9h-19h	2+2
N_RUR_3	1999 (with recent renovations)	CR (4Y)	4Y	50.2	23	9h-17h30	2+0
		CR (5Y)	5Y		17	9h-17h30	2+2
PRIM_RUR_1	2011	CR (6Y-1st grade)	1st	46.9	19	9h-12h, 13h30-17h30	2+0
		CR (8Y-3rd grade)	3rd		26		2+2
		LR[2]	LR		160	12h-13h30	2+0

[1] CR – classroom; [2] LR - lunch room

Table 2. Statistical parameters of the hourly mean data for each microenvironment studied.

	School	N_RUR_1	N_RUR_2		N_RUR_3		PRIM_RUR_1		
	Room	3-5Y	0Y	2Y	4Y	5Y	1st	3rd	LR
Radon (Bq m^{-3})	Min	58.42	32.13	0.00	99.32	67	105.16	242.45	2.92
	Max	359.29	157.74	108.08	277.50	386	344.68	888.00	292.11
	Mean	222.64	100.58	38.53	167.96	206	242.03	632.68	143.07
	Median	227.84	102.24	35.05	166.50	199	242.45	660.16	141.67
T (ºC)	Min	19	-	20	19	16	17	17	16
	Max	22	-	26	23	25	21	24	21
	Mean	20	-	23	21	19	19	19	18
	Median	20	-	23	21	19	18	19	17
RH (%)	Min	36	-	23	36	36	51	40	34
	Max	54	-	47	59	52	70	62	57
	Mean	43	-	23	45	44	58	52	53
	Median	42	-	23	44	46	58	53	54

[1] CR – classroom; [2] LR - lunch room

Table 3. Exceedances (%) to the reference values during occupation periods.

Building	Room	Non-compliances for T (%)	Non-compliances for RH (%)	Radon exceedances (%)				
		ASHRAE[a]	ASHRAE[b]	USEPA[c]	WHO[d]	WHO[e]	EU[f]	PT[g]
N_RUR_1	3-5Y	4	0	39	83	0	0	0
N_RUR_2	0Y	-	-	14	57	0	0	0
	2Y	48	0	0	0	0	0	0
N_RUR_3	4Y	0	0	67	94	0	0	0
	5Y	22	0	85	100	7	0	0
PRIM_RUR_1	1st	45	9	82	100	0	0	0
	3rd	19	0	100	100	100	88	100
	LR	50	0	0	0	0	0	-

[a] 20-23.9 ºC (winter); [b] 30-60%; [c] 150 Bq/ m^3; [d] 100 Bq/ m^3; [e] 300 Bq/ m^3; [f] 400 Bq/ m^3; [g] Portuguese legislation-400 Bq/ m^3 (8 hr mean)