

RADON CONCENTRATIONS ON THE NUCLEAR AND RADIOACTIVE INSTALATIONS OF NUCLEAR REACTOR CENTER – CRPQ/IPEN

Paulo Sergio Cardoso da Silva, Marcia Pires de Campos and Guilherme de Lima Reis

Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
pscsilva@ipen.br
mpecampos@ipen.br
glreis@ipen.br

ABSTRACT

Nuclear and radioactive workers are normally exposed to dose resulting from their day by day activities. Besides that, the ubiquitous radon distribution can also contribute for the exposure rates. The radionuclide ^{222}Rn is a noble gas belonging to the uranium series and its indoor concentration in the air depend on exhalation from surrounding soil and on exhalation from building materials. Radon emanating from porous building materials may achieve large relevance in areas with high uranium concentrations and areas with limited ventilation. The objective of this study was to evaluate the ^{222}Rn concentrations in the radiochemistry and radiometric laboratories and in the reactor nuclear building of the Nuclear Reactor Center (CERPq) located in the Nuclear and Energy Research Institute (IPEN). Measurements were done by using a Radon Gas Monitor, model RAD7, produced by Durrige Company equipped with a solid-state alpha detector and a passive method, with SSNTDs placed within small diffusion chambers, as detectors square pieces (2.5 cm \times 2.5 cm) of CR-39 foils were used. The CR-39 detectors were etched in KOH 30% solution at 80 °C for 5.5 h in a constant-temperature bath. After etching, the detectors were washed, dried, and scanned under a Carl Zeiss microscope to obtain the track density measurements. The activity concentrations varied from 52 to 103 Bq m⁻³ for the measured areas in CERPq. These values are in accordance with what is established by the World Health Organization for safe environments of 100 Bq m⁻³.

1. INTRODUCTION

The main source of ionizing radiation to the population exposure comes from natural radioactivity and radon and its progenies contribute with more than 50% to annual effective dose received considering all-natural sources of ionizing radiation [1]. Radon is a naturally occurring, colorless and odorless radioactive gas. The most frequent and relevant isotope from an epidemiological point of view is Radon-222, with a half-life of 3.8 days, and it tend to accumulate indoors, particularly in places with low ventilation. When inhaled, the radioactive decay products of radon may cause irradiation of the lung, mainly the short-live descendants ^{218}Po and ^{214}Po . According to the World Health Organization's International Agency for Research on Cancer (IARC) radon is classified as a group 1 carcinogen [2, 3].

The World Health Organization [4] establishing in 2009 a recommended action level for residential radon of 100 Bq/m³ and 300 Bq/m³ as the concentration that must not be surpassed in any case. Indoor radon exposure includes not just dwellings, but also

workplaces. Workplaces that can be considered radon-prone areas include underground work and some specific industries.

Worldwide different levels are adopted as reference levels. The International Commission on Radiological Protection (ICRP) [5] suggests the action level for non-related nuclear and radioactive workers values varying from 500 to 1500 Bq m⁻³, considering an equilibrium factor of 0.4 and dose conversion conventions. The Health and Safety Executive, of United Kingdom adopts a radon action level of 400 Bq m⁻³ for workplaces based on advice from the National Radiological Protection Board (NRPB) [6]. Other countries establish lower levels, as for example, Estonia [7] and the United States, where reference levels are 200 Bq m⁻³ and 150 Bq m⁻³, respectively.

Nuclear and radiation-related works may be exposed to higher doses in the daily execution of their activities, reason why the normal exposure to these individuals is 20 mSv/yr while for public individuals, the dose increment is limited to 1 mSv/yr [8]. Although the general contribution of Rn-isotopes, that arises from the nuclear energy production, is lower than 1% of the total dose (UNSCEAR, 2010) (the combination of all natural and anthropogenic sources), there are few data on the radon levels in nuclear and radioactive workplaces other than uranium mining [9, 10, 11].

The objective of this paper was to describe the radon concentrations in the radioactive laboratories and nuclear installations of the Centro do Reator de Pesquisa – CERPq, a unit of the Instituto de Pesquisas Energéticas e Nucleares – IPEN, located in the Cidade Universitária, São Paulo.

2. METHODOLOGY

The CERPq houses the nuclear research reactor IEA-R1, radiochemical and radiometric laboratories. The IEA-R1 reactor that operates in the radioisotope production, neutron irradiation for neutron activation analysis and material characterization, most of them performed in the CERPq installations. A simplified scheme of the center is showed in Figure 1. Radon measurements were performed in buildings 2 and 3.

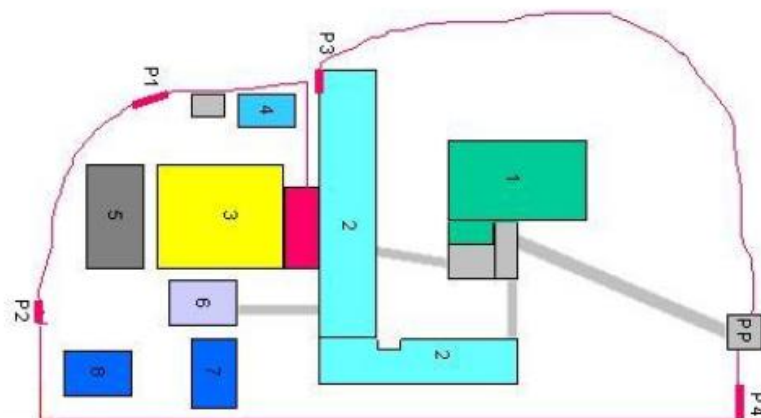


Figure 1: Simplified scheme of the CERPq buildings. 2 - radiochemical and radiometric laboratories, 3 - IEA-R1 reactor.

The IEA-R1 is an open pool type reactor, installed in an all-closed building with thick walls and controlled ventilation system to avoid the air flowing to the outside. In this building two ^{222}Rn measurements were done in the hall of the reactor pool (R1 and R2) and one measurement in the nuclear physics laboratory installed in first floor of the reactor building (FF) for conducting experiments demanding neutron flux. The radiometric laboratories are used for gamma counting of the irradiated sample for neutron activation analysis. One measurement was done in the rooms 126, 128 and 130, located in the building 2 of Figure 1. In these rooms the air is controlled by a central air conditioning system and the doors remain closed most of the time. The radiochemical laboratories, also located in building 2, number 134, 135, 136, 137 and 226 are used mainly for sample preparation for neutron activation analysis before irradiation and for counting, after. Room 139 is used as a deposit. Some of these rooms have simple air conditioning system that are not continuously turned on. The windows of all the laboratories remains closed almost the entire time.

2.1 Radon Measurements

Radon measurements in the air of the laboratories and in the pool hall of the reactor were done by using two different methodologies: RAD7 and solid-state nuclear trace detectors (SSNTD).

2.1.1. RAD7

Measurements of radon used a RAD7 (Model: RAD7, DurrIDGE, US) solid state ion implanted silicon alpha detector. The detector converts alpha radiation directly to an electric signal allowing to discriminate alpha particles by energy. The radon activity concentrations are determined by the decay of ^{218}Po and ^{214}Po trapped inside a chamber cell of 0.7 L. A calibration accuracy of the instrument is guaranteed by the manufacturer with a calibration precision better than 5% [12].

2.1.2. SSNTD

Nuclear solid-state trace detectors (SSNTDs) are solid material that present grooves on its surface when exposed to a defined radiation. Among the commercially available trace detectors, the CR-39 was used because it presents better optical quality, since it is transparent, and it allows a contrast between the produced trace and the plastic body itself. After exposure the detectors underwent chemical attack with potassium hydroxide solution, 30% (w/v), for 5.5 hours at 80 °C. The traces in the detector were observed using a ZEISS Axiolmager light microscope for transmitted light model with increase of 10x. The equipment was connected to a video camera Zeiss ICC-1, connected to a microcomputer with an HP 29" monitor.

Rn-222 concentration was calculated according to equation 1 [13], taking the density of traces (traces/cm²), the exposure time and the calibration factor, that relates the trace density in the surface of the detector and the concentration of radon.

$$C_{Rn} = \frac{D}{kxt} \quad [1]$$

At where:

C_{Rn} = ²²²Rn concentration (Bq/m³);

k = calibration factor (traces/cm² per Bq/m³ d⁻¹);

D = Net trace density (discounting the density relative to background radiation in the detector) (traces/cm²);

t = exposure time (d);

3. RESULTS AND DISCUSSION

The results for RAD7 measurements are shown in Figures 2 for the radiochemical laboratories, in Figure 3 for the radiometric laboratories and in Figure 4 for the hall of the reactor pool and in the laboratory of nuclear physics in the first floor of the building. RAD 7 also provides information on the temperature and humidity and the results, together with the ²²²Rn concentrations are summarized in Table 1.

Even without a strict control, the temperature and humidity of the radiochemical laboratories do not vary in a great extension. The variation of temperature is lower than 6% and for the humidity, lower than 5% for all the radiochemical laboratories. The mean values of ²²²Rn concentration in the air of these laboratories varied from 23 to 42 Bq m⁻³. The highest values for ²²²Rn concentration was observed in room 135, reaching out 82 Bq m⁻³.

In the radiometric laboratories, temperature is controlled by a central air conditioning system. The mean values varied from 19.4 to 21.4 °C for the three rooms, lower than mean temperature of the radiochemical laboratories that varied from 22.3 to 25.3 °C. Humidity was lower in the radiological laboratories (varying from 48.2 to 63.2%) than in radiochemical ones (variation of 52.8 to 69.9%). The mean values of ²²²Rn concentration varied from 36 to 43 Bq m⁻³ with the higher values observed in the room 126, reaching 89 Bq m⁻³.

Table 1: Minimum, maximum, mean values and standard deviation of ^{222}Rn activity concentration, temperature and humidity in the CRPq

	Radiochemical labs					Radiometric labs				Reactor		
	134	135	136	137	139	226	126	128	130	R1	R2	FF
Concentration (Bq/m^3)												
min	15	11	8	9	13	9	8	20	24	16	30	30
max	56	82	68	57	65	51	89	65	74	81	74	83
mean	38	42	23	26	34	31	38	36	43	48	49	51
SD	11	16	11	10	12	9	16	10	11	10	9	9
Temperature ($^{\circ}\text{C}$)												
min	23.1	21.9	22.8	21.9	20.7	21.9	18.2	17.3	19.7	22.5	23.1	20
max	24.9	28.3	26.4	23.1	24.9	26.8	26.4	21.6	24.9	30.1	31.3	29.2
mean	23.7	25.3	24.7	22.4	22.9	24.3	21.4	19.4	21.4	27.0	28.4	26.5
SD	0.4	1.5	0.7	0.4	1.3	1.4	2.7	1.1	1.0	2.0	2.2	1.7
Humidity (%)												
min	60	48	48	65	60	58	48	52	41	49	52	51
max	68	59	60	74	74	70	67	74	53	78	78	74
mean	64.3	52.8	53.4	69.9	68.0	64.0	58.3	63.2	48.2	68.1	67.6	64.8
SD	1.5	2.3	2.1	2.1	2.4	3.2	4.8	5.5	2.9	7.6	6.7	6.0

The hall of the reactor pool (R1 and R2) and the physics laboratory of the first floor (FF) presented temperature and humidity variations lower than 7.8% and 11.1%, respectively. The mean values of ^{222}Rn concentrations varied from 48 to 51 Bq m^{-3} and the highest values was observed reached 83 Bq m^{-3} in the first floor.

The results of the radon concentration measured with CR-39 are shown in Table 2, together with the mean concentration obtained with RAD7 detector, for comparison. Generally, the concentrations obtained with CR-39 are higher than that obtained with RAD7. Results obtained with RAD7 are mean values of measurements taken each five hours, for one week, while the ones obtained with CR-39 are the integrated value covering a period of three months. This difference in the time of data collection can be the responsible for the observed differences. Espinosa et al. [14], in an intercomparing of indoor radon measurement using nuclear track detectors and different dynamic recording systems, also found lower radon concentrations for RAD7 compared to CR-39. The authors explained that the lower values can be due the presence of an integrated humidity filter in the RAD7 and ionized radon atoms may adhere to water molecules present in the cellar atmosphere, being retained by the filter [15].

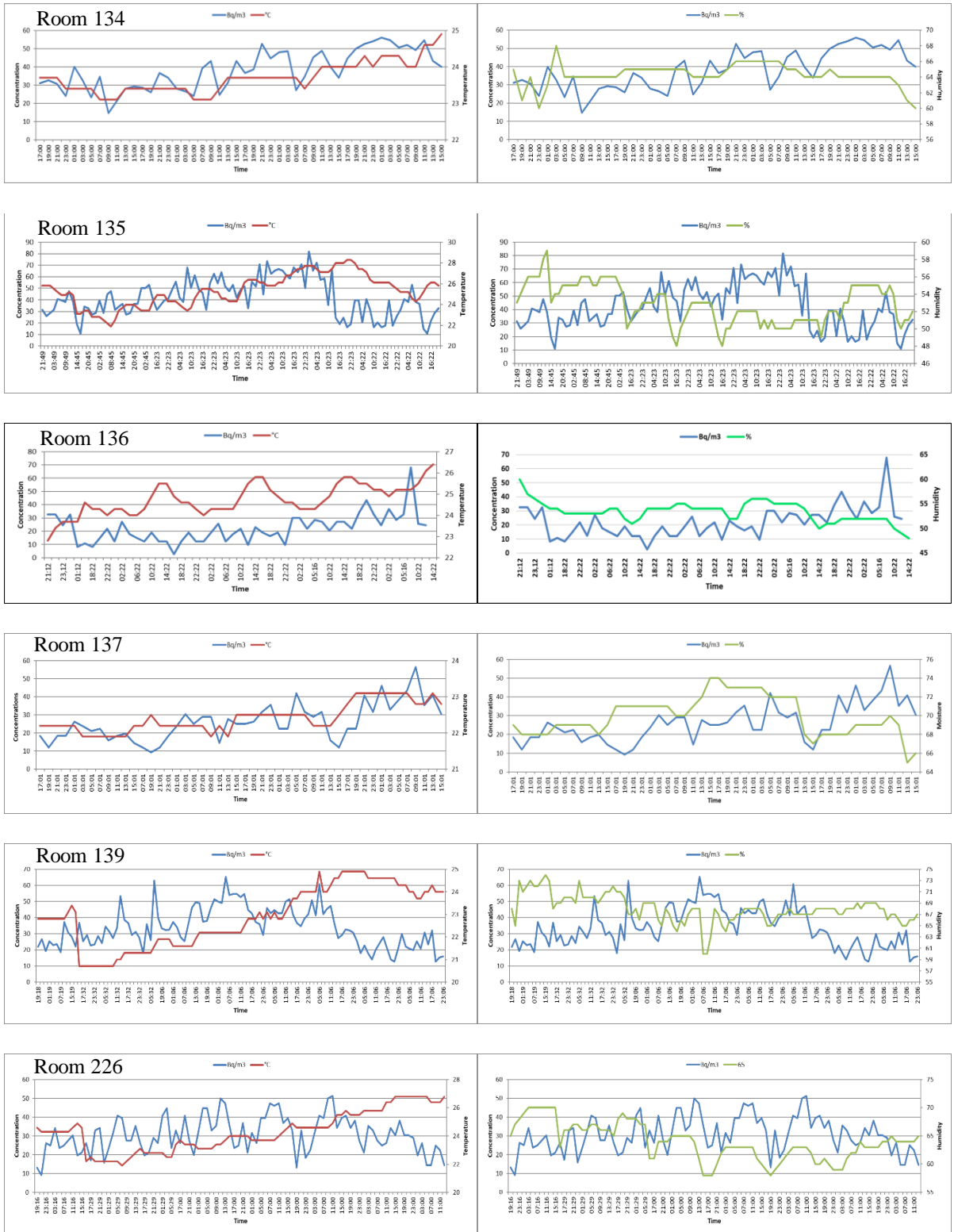


Figure 2: Radon concentration (Bq m^{-3}), temperature ($^{\circ}\text{C}$) and ai humidity (%) in the radiochemical laboratories.

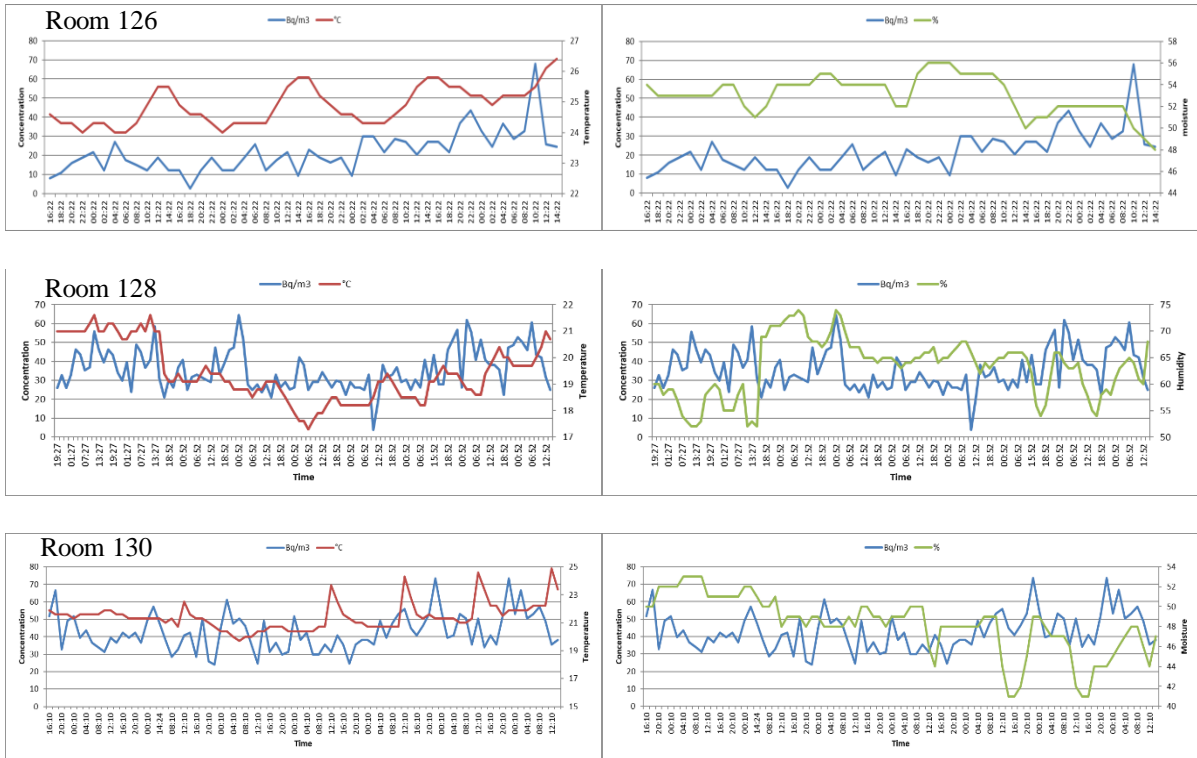


Figure 3: Radon concentration (Bq m^{-3}), temperature ($^{\circ}\text{C}$) and ai humidity (%) in the radiometric laboratories.

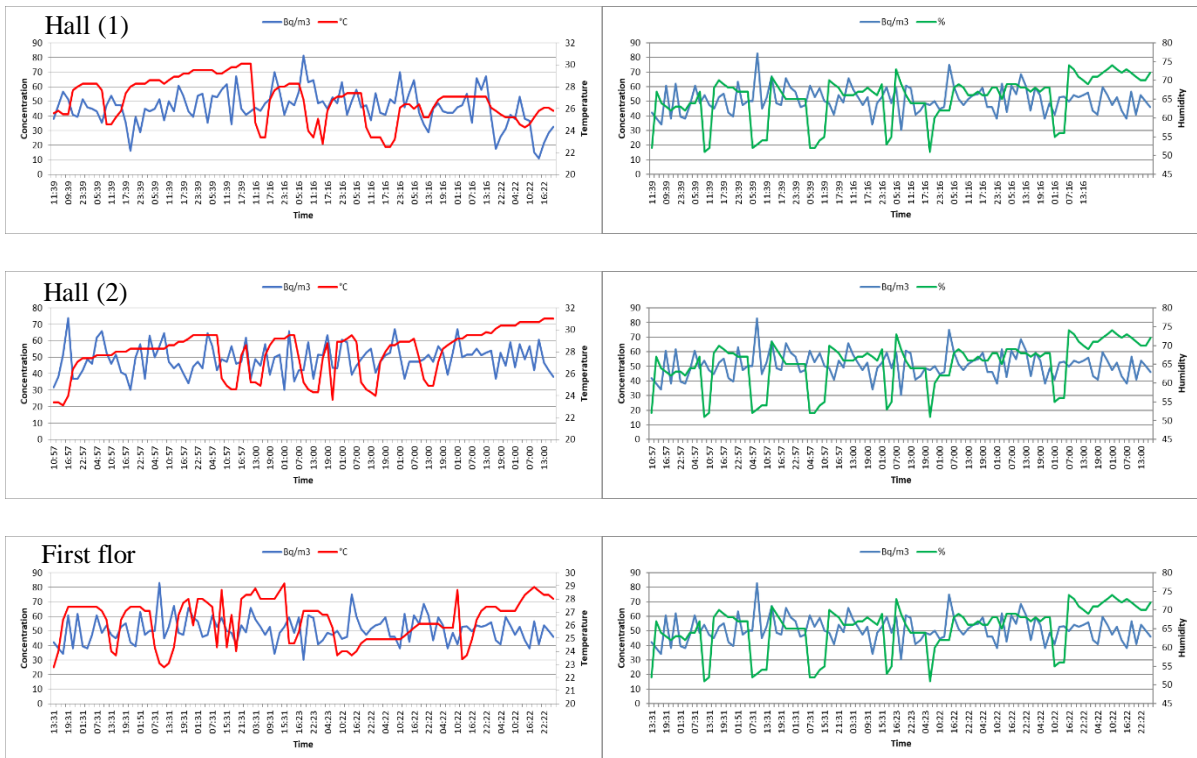


Figure 4: Radon concentration (Bq m^{-3}), temperature ($^{\circ}\text{C}$) and ai humidity (%) in the hall of the reactor pool (two measurements (1) and (2)) and the laboratory of the reactor first floor.

Table 2: Radon concentration (Bq m^{-3}) obtained by RAD7 and CR-39 and dose assessment (mSv a^{-1}) for the laboratories and reactor hall of CRPq

	C (Bq/m^3)		E (mSv a^{-1})
	RAD7	CR-39	
Radiochemical labs			
134	38	74	0,44
135	42	52	0,31
136	23	59	0,35
137	26	54	0,32
139	34	66	0,39
226	31	63	0,38
Radiometric labs			
126	38	67	0,40
128	36	60	0,36
130	43	72	0,43
Reactor			
R1	48	90	0,54
R1	49	102	0,61
FF	51	103	0,72

Table 3 shows a compilation of ^{222}Rn activity concentrations in dwellings, workplaces and underground workplaces in different parts of the world for comparison. Despite the differences in the two measurements methods, the ^{222}Rn concentration determined in the laboratories of the CERPq and in the hall of the reactor pool are in the same range as that observed for dwellings and workplaces and are much lower than radon concentrations measured in underground workplaces and caves. Also, the values are below or equal the action level for residential radon of 100 Bq/m^3 recommended by the World Health Organization (WHO, 2009), ICRP 65, Health and Safety Executive of United Kingdom, and US levels, however the measurement technique used.

Figure 5 shows that the concentrations obtained by RAD7 and CR-39 measurements present a good correlation with correlation coefficient of 0.79 being the first approximately 50% lower than the second.

The effective dose for radon inhalation is also presented in Table 2. The higher doses are observed for the reactor pool area and the laboratory located in the first floor of reactor building. According to UNSCEAR 2000 the average worldwide effective dose due to radon inhalation is 1.26 mSv/y . The radon concentrations found for all CERPq areas, both radioactive and nuclear, are below this value.

Besides the rooms measured with both methodologies, two others were measured only with CR-39 track detector: rooms 132 and 133. The first one is used as a place for decaying of the irradiated samples and the second is used to open the aluminum capsules inside with the samples are irradiated. The radon concentrations were 55 and 56 Bq m^{-3} , respectively and the effective dose were 0.33 mSv/y for both rooms.

Table 3: Radon concentration (Bq m^{-3}) found in residences, workplaces, underground and caves

		Mean	Range		
Palakkad, India	Dwelling	28.1	15 - 79	[16]	2017
Al-kharj, Saudi Arabia	Dwelling	114	67 - 488	[17]	2014
	Workplace	76	46 - 267	[17]	
South- Day, Gana	Indoor	24.9	27 - 42.8	[18]	2018
Eastern Sicily, Italy	Indoor	53	24 - 126	[19]	2012
Brisbane, Australia	Workplace	10.5	0.7 - 86.6	[20]	2015
Catalonia, Spain	Workplace, Underground		< 1 – 12.900	[21]	2008
Mexico City, Mexico	Dwelling	28		[22]	2009
	Workplace	123		[22]	
Guadalajara	Dwelling	80		[22]	
	Workplace	160		[22]	
Monterrey	Dwelling	42		[22]	
	Workplace	69		[22]	
Stan Ter, Kosovo	Workplace, underground	281.4	60 – 748	[22]	
Italy	Workplace, underground	723	7 – 43.919	[23]	2009
São Paulo, Brazil	Caves (PETAR)		515 – 6.607	[24]	2005

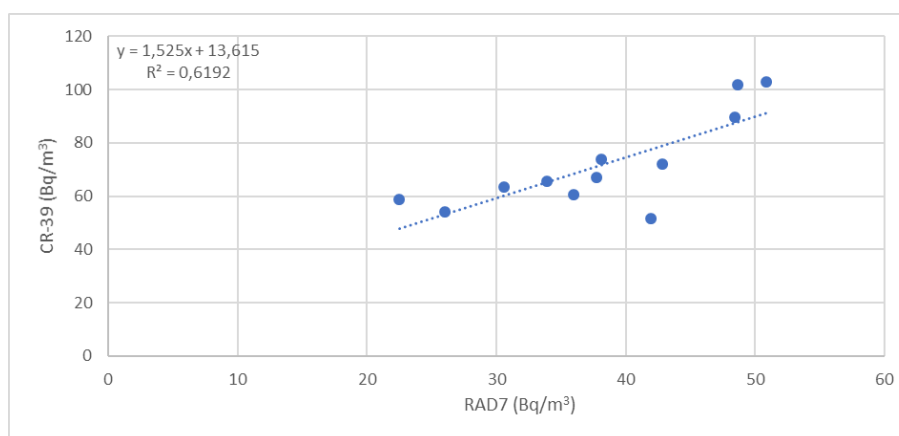


Figure 5: Linear regression obtained for radon concentration (Bq m^{-3}) measured by RAD7 and CR-39.

3. CONCLUSIONS

The control of radon exposition is of prime importance for the population in general and for radioactive and nuclear workplaces, in particular, since these places offers a higher risk of radiation exposition by the very nature of the activity. Radon concentrations were measured in radiochemical and radiometric laboratories, in the hall of the reactor pool, and in a physics

laboratory installed in the same building of the IEA-R1 reactor at CERPq using an active alpha particle detector (RAD7) and the passive CR-39 detector. Mean concentration values were in the range of 31 to 51 Bq m⁻³, with RAD7 measurements and 52 to 103 Bq m⁻³, with CR-39 measurements. These concentrations are in the same range as the ones observed for dwelling and non-radioactive and non-nuclear workplaces and are lower than the concentrations that can be found in underground and caves. The annual effective doses are in the range of 0.31 to 0.72 mSv, being the higher observed values almost 50% of the worldwide effective dose due to radon inhalation.

ACKNOWLEDGMENTS

Authors are thankful to Fapesp for the grants number 2003/08146-2 and 2006/01112-3 and for the CERPq staff that contributed with the research.

REFERENCES

1. UNSCEAR. Sources and Effects of Ionizing Radiation. Volume I: Sources: Report to the General Assembly, Scientific Annexes A and B. UNSCEAR 2008 Report. United Nations Scientific Committee on the Effects of Atomic Radiation. United Nations sales publication E.10.XI.3. United Nations, New York, (2010).
2. IARC, IARC monographs on the evaluation of the carcinogenic risk to humans. In: Man-made Fibres and Radon, (1998).
3. IARC, IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, vol. 100D Radiation, (2012).
4. World Health Organization. WHO Handbook on indoor radon: a public health perspective. Geneva, Switzerland: World Health Organization; pp 94, (2009).
5. ICRP, International Commission on Radiological Protection, Protection against radon-222 at home and at work ICRP Publication 65; Ann. ICRP 23, (1993)
6. D. W. Dixon, T. D. Gooding, S. McCready-Shea, Evaluation and significance of radon exposures in British workplace buildings, *Environment International*, **22**, pp. S1079-S1082 (1996).
7. L. Pahapill, A. Rulkov, R. Rajamäe, G. Åkerblom, Radon in Estonian dwellings. Results from a National Radon Survey. SSI rapport, 16, pp 20, (2003).
8. Resolução CNEN-114/2011 —Diretrizes Básicas de Proteção Radiológica, de 01 de setembro de 2011. Norma CNEN-NN-3.01.
9. K. Matshusa, M. Makgae, Prevention of future legacy sites in uranium mining and processing: The South African perspective, *Ore Geology Reviews*, **86**, pp 70-78, (2017).
10. Q. Zhou, S. Liu, L. Xu, H. Zhang, D. Xiao, J. Deng, Z. Pan, Estimation of radon release rate for an underground uranium mine ventilation shaft in China and radon distribution characteristics, *Journal of Environmental Radioactivity*, **198**, pp 18-26, (2019).
11. K. Ajayi, K. Shahbazi, P. Tukkaraja, K. Katzenstein, “Numerical investigation of the effectiveness of radon control measures in cave mines”, *International Journal of Mining Science and Technology*, **29** (3), pp 469-475 (2019).
12. Durridge, 2019, RAD7 Calibration Services, <https://www.environmental-expert.com/services/rad7-calibration-services-233942>, Accessed in 6/07/2019.

13. Y. S. Mayya, K. P. Eappen, K. S. V. Nambi, "Methodology for mixed field inhalation dosimetry in monazite areas using a twin-cup dosimeter with three track detectors", *Rad. Prot. Dosim.*, **77(3)**, pp 177-184 (1998).
14. G. Espinosa, J. I. Golzarri, M. I. Gaso, M. Mena, N. Segovia, "An intercomparison of indoor radon data using NTD and different dynamic recording systems", *Radiation Measurements*, **50**, pp 112-115 (2013).
15. M. Cerda-Zorrilla, Indoor radon mitigation by means of water capture. thesis, Universidad Nacional Autonoma de México. (2012).
16. M. Ramsiya, Antony Joseph, P.J. Jojo, "Estimation of indoor radon and thoron in dwellings of Palakkad, Kerala, India using solid state nuclear track detectors", *Journal of Radiation Research and Applied Sciences*, **10:3**, pp 269-272 (2017).
17. A. M. Maghraby, K. Alzimami, M. Abo-Elmagd, "Estimation of the residential radon levels and the population annual effective dose in dwellings of Al-kharj, Saudi Arabia", *Journal of Radiation Research and Applied Sciences*, **7 (4)**, pp 577-582 (2014).
18. C. Y. Ansre, M. K. Miyittah, A. B. Andam, D. E. Dodor, "Risk assessment of radon in the South Dayi District of the Volta Region, Ghana", *Journal of Radiation Research and Applied Sciences*, **11(1)**, pp 10-17 (2018).
19. R. Catalano, G. Immè, G. Mangano, D. Morelli, A. R. Tazzer, "Indoor radon survey in Eastern Sicily", *Radiation Measurements*, **47(1)**, pp 105-110 (2012).
20. S. H. Alharbi, R. A. Akber, "Radon and thoron concentrations in public workplaces in Brisbane, Australia", *Journal of Environmental Radioactivity*, **144**, pp 69-76 (2015).
21. L. I. Font, C. Baixeras, V. Moreno, "Indoor radon levels in underground workplaces of Catalonia, Spain", *Radiation Measurements*, **43 (1)**, pp S467-S470, (2008).
22. G. Espinosa, J. I. Golzarri, A. Angeles, R. V. Griffith, "Nationwide survey of radon levels in indoor workplaces in Mexico using Nuclear Track Methodology", *Radiation Measurements*, **44 (9-10)**, pp 1051-1054 (2009).
23. V. Carelli, V. Bianco, C. Cordedda, G. Ferrigno, C. Carpentieri, F. Bochicchio, "A national survey on radon concentration in underground inspection rooms and in buildings of a telephone company: methods and first results", *Radiation Measurements*, **44 (9-10)**, pp 1058-1063 (2009).
24. S. Albergi, B. R. S. Pecequilo, M. P. Campos, "Radon concentrations in caves of Parque Estadual do Alto Ribeira (PETAR), SP, Brazil: preliminary results", *International Congress Series*, **1276**, pp 403-404 (2005).