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2014 J. Radiol. Prot. 34 339

(http://iopscience.iop.org/0952-4746/34/2/339)

IOP Publishing | Society for Radiological Protection Journal of Radiological Protection

J. Radiol. Prot. **34** (2014) 339–346 doi:10.1088/0952-4746/34/2/339

Radon exposure at a radioactive waste storage facility

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Received 25 September 2013, revised 13 February 2014
Accepted for publication 14 February 2014
Published 4 April 2014

Abstract

The Waste Management Department of Nuclear and Energy Research Institute (IPEN) is responsible for the safety management of the waste generated at all internal research centers and that of other waste producers such as industry, medical facilities, and universities in Brazil. These waste materials, after treatment, are placed in an interim storage facility. Among them are ^{226}Ra needles used in radiotherapy, siliceous cake arising from conversion processes, and several other classes of waste from the nuclear fuel cycle, which contain Ra-226 producing ^{222}Rn gas daughter.

In order to estimate the effective dose for workers due to radon inhalation, the radon concentration at the storage facility has been assessed within this study. Radon measurements have been carried out through the passive method with solid-state nuclear track detectors (CR-39) over a period of nine months, changing detectors every month in order to determine the long-term average levels of indoor radon concentrations. The radon concentration results, covering the period from June 2012 to March 2013, varied from 0.55 ± 0.05 to 5.19 ± 0.45 kBq m^{-3}. The effective dose due to ^{222}Rn inhalation was further assessed following ICRP Publication 65.

Keywords: radon, solid-state nuclear track detectors, waste

1. Introduction

Radioactive waste is generated in practices that use radioactive materials during relevant operational processes. These practices include all uses of radionuclides in industry, medicine, and research [1, 2]. After generation, and before transportation to final disposal, untreated radioactive waste must be treated and conditioned. After treatment, radioactive waste may be subject to interim storage for varying periods of time. The interim storage space can be located

0952-4746/14/020339+08$33.00

Table 1. Characteristics of the packages at the interim storage facility.

Waste class	Package type	Package quantity	Waste volume (m^3)	Activity (GBq)	Major isotopes (% estimated activity)
Compactable	200 l drum	813	162.6	6.9×10^2	NDC[b](20), Others (80)
Not compactable	200 l drum	417	83.4	6.9×10^1	NDC (80), Others (20)
	1.5 m^3 box	1	1.5	1.0×10^{-1}	NDC (80), Others (20)
Biological	200 l drum	2	0.4	CNC[a]	^{14}C (100)
Ash	200 l drum	9	1.8	5.0×10^{-3}	^{232}Th (100)
Cake	200 l drum	33	6.6	1.1×10^{-1}	^{238}U (80), ^{232}Th (20)
Sealed source	200 l drum	12	2.4	2.6×10^3	^{60}Co (53), ^{137}Cs (42), Others (5)
Radium	200 l drum	25	12.5	5.4×10^2	^{226}Ra (100)
Bale	1.5 m^3 box	50	75.0	1.7×10^2	^{137}Cs (100)

[a] Characterization not concluded.

[b] Natural decay chains, mainly ^{238}U and ^{232}Th.

at the same place where the waste was generated, or it can be located in a centralized facility [3]. The interim storage facilities must comply with international recommendations and national regulations of the regulatory body [4, 5].

The Waste Management Department (GRR) of the Nuclear and Energy Research Institute—Instituto de Pesquisas Energéticas e Nucleares, São Paulo (IPEN/SP) is responsible for receiving, processing, and storing the radioactive waste generated at research centers and other non-nuclear installations in Brazil, according to Brazil's Federal Law 7781 published in 1989. It was decided in 2010 that all treated radioactive waste packages, already at various interim storage facilities, had to be transferred to a new interim storage facility. This new facility is a 17 m long by 25 m wide by 6 m high building of total volume equal to 2550 m^3, with a single door and no windows or artificial ventilation.

The stored packages within this new facility are classified according to geometry and content as compactable, not compactable, biological, ash, cake, sealed sources, radium, and bale. A brief description of each class is presented below; some other package characteristics are presented in table 1.

- Compactable—small objects, tissues, swabs, paper, cardboard, plastics (polyvinylchloride, polyethylene), rubber gloves, protective clothes, and glassware treated by compaction in a 10 ton press in 200 l drums.
- Not compactable—metallic scrap, wood, brickwork, filters, charcoal, tools, and parts of equipment. This waste is conditioned either in 200 l drums or in 1.5 m^3 boxes.
- Biological—animal carcasses treated by mummification with CaO in 200 l drums.
- Ash—ash arising from a fire accident in a factory of gas lamp mantles, immobilized in cement paste in 200 l drums.
- Cake—siliceous cake arising from the dissolution step of the uranium conversion process, immobilized in cement paste in 200 l drums.
- Sealed sources—spent sealed sources either inside the original shielding or transferred to a standard shielding, encapsulated in concrete in 200 l drums.
- Radium—radium needles conditioned inside a sealed metallic container and encapsulated in concrete in 200 l drums.
- Bale—paper bales from the Goiânia radiological accident, conditioned in 1.5 m^3 boxes.

Surface contamination by α, β and γ emitters was detected in mid-2010 on the shoes of workers in the storage facility, and some wipe tests indicated similar contamination on the surface of the packages. It was observed that the presence of these contaminants on the wipes disappeared after a period of around 4 h, behavior very similar to that of ^{222}Rn daughters, i.e., the isotopes ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po [6]. Following this, the Environmental Radiometry Laboratory of IPEN/SP was assigned to investigate the ^{222}Rn levels in the air inside this interim storage facility.

It is known that ^{222}Rn is an inert gas that may become airborne by diffusing into the air. The main health hazard associated with ^{222}Rn is due to its short-lived daughter products, many of which are alpha emitters causing increased lung exposure. Epidemiological studies show the correlation between exposure to radiation and excess lung cancer and reveal that radon exposure is the one of the main causes of this cancer after smoking [7, 8].

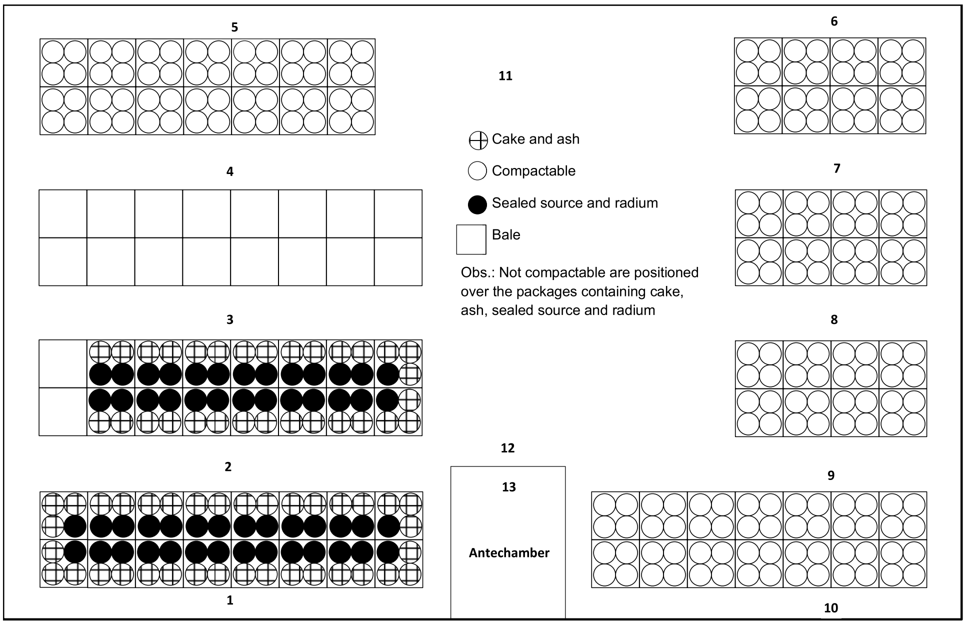
Preliminary results of radon concentration levels at the storage facility indicated concentrations exceeding 1000 Bq m^{-3}, which is the action level for applying occupational protection measures for existing exposure situations [9]. Based on these results, research was started aiming at the accurate determination of radon levels inside the interim storage facility of the GRR of IPEN/SP. Radon measurements were carried out through the passive method, using solid-state nuclear track detectors (SSNTDs) of the CR-39 type, over a period of nine months. The detectors were changed every month, in order to determine the long-term average levels of the indoor radon concentrations. The effective dose due to radon inhalation for workers was obtained from the measured radon concentrations using the dose conversion factor according to International Commission on Radiological Protection (ICRP) Publication 65 [7].

A review in the literature reveals that there are studies related to radon concentration levels in final disposal facilities [10, 11] and also related to pre-treatment processes [12], but there are no specific studies regarding ^{222}Rn concentration measurements in interim storage facilities.

2. Materials and methods

A wide variety of well-established techniques is available for the measurement of radon levels [13]. The passive method, with SSNTDs placed within small diffusion chambers, has been used widely, since the detectors are relatively inexpensive, reliable, and generally unaffected by climatic conditions variability [14–16]. In this study, the ^{222}Rn concentration was measured by this particular passive detection method. Square pieces (2.5 cm × 2.5 cm) of CR-39 foils were used as detectors. A hemispherical 'close-can' of 4 cm radius was selected as the diffusion chamber (National Radiological Protection Board or NRPB dosimeter, now called the Radiation Protection Division of the Health Protection Agency) [15]. This chamber consist of a polypropylene holder made of an upper and lower half which snap together during assembly. The fitting of the two halves is quite tight, in order to prevent dust and radon progeny entry and access of moisture. Thoron is not diffused into the chamber due to its short half-life and diffusion time. The detectors were suspended in the facility from the ceiling at a height of approximately 1.7 m from the ground and placed at a distance from any surface, to avoid the pleat-out effect. After exposure for approximately 30 days, the detectors were collected and replaced by new ones.

All detectors were etched in KOH (30% mass weight) solution at 80 °C for 5.5 h in a constant-temperature bath [17]. After etching, the detectors were washed, dried, and scanned under a Carl Zeiss microscope to obtain the track density measurements. The background was 30 ± 2 tracks cm^{-2}. Using a calibration factor of 0.0505 ± 0.0051 tracks cm^{-2} per Bq m^{-3} d, obtained with a Pylon model RN-150 calibrated radon gas source, the track density



Obs.: Not compactable are positioned over the packages containing cake, ash, sealed source and radium

Figure 1. Sampling points inside the investigated radioactive waste storage facility.

was converted to radon concentration in the environment. The Pylon Type RN-150 calibrated radon gas source provides dry radon gas (^{222}Rn) for calibration of both scintillation cells and radon gas measurement equipment [17, 18].

The radon concentrations were calculated through the following equation [19]:

$$C = \frac{D}{kt} \tag{1}$$

where C is the radon concentration (Bq m^{-3}), D is the track density (tr cm^{-2}), k is the calibration factor (tr cm^{-2} per Bq m^{-3} d), and t is the exposure time (d).

In total, 13 sampling points were set up; 12 of them were located inside the facility and the last one was located in the antechamber (figure 1). In order to obtain radon concentration measurements with better statistical significance, two diffusion chambers were installed at each sampling point.

In total, seven sampling campaigns were conducted from June 2012 to March 2013; each of them had a mean duration of 30 days.

3. Results and discussion

3.1. Radon concentration

The average radon concentration results at the radioactive waste storage facility from June 2012 to March 2013 are presented in table 2. The average radon concentration obtained from the current study varied from 0.55 ± 0.05 to 5.19 ± 0.45 kBq m^{-3} inside the radioactive waste storage facility. In the antechamber, the values varied from 0.31 ± 0.02 to 0.95 ± 0.06 kBq m^{-3}. The minimum values of the average radon concentration were recorded in the first sampling campaign from June to August 2012, whereas the maximum values were found in the period

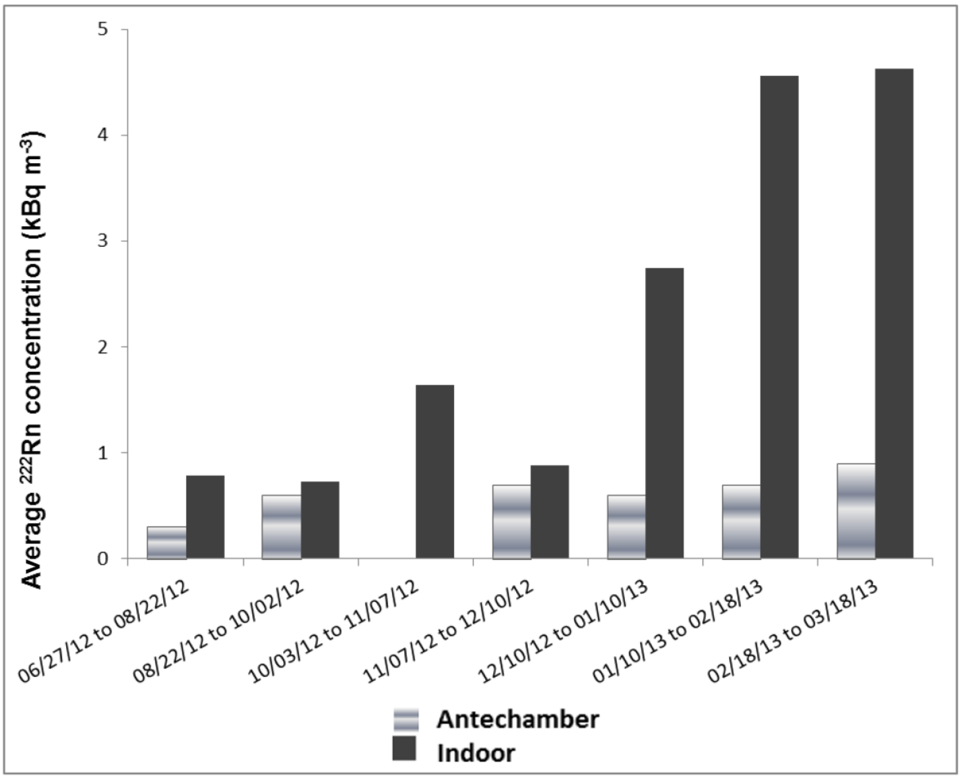


Figure 2. Average ^{222}Rn concentration in the investigated radioactive waste storage facility from June 2012 to March 2013.

from February to March 2013. According to the results presented in table 2, there was no significant variation of radon concentration among the 12 points monitored inside the facility for a given sampling campaign. So, it was not possible to identify any major source of radon. To get a clear picture of the average radon levels at the radioactive waste storage facility, a graph was plotted; see figure 2.

As can be seen in figure 2, there is a wide variation in average radon concentration, and some results exceeded the action level of 1000 Bq m^{-3} [9]. It should be pointed out that during the first, second, and fourth sampling campaigns there was intense labor activity at the radioactive waste storage facility and the door remained continuously open, resulting in a considerable rate of air exchange. From December 2012 to March 2013, the facility remained closed most of the time, having been opened only for the exchange of detectors. As previously mentioned, there are no windows at the facility. So, a plausible explanation for the variation of average radon concentration during the sampling campaigns would be the different conditions of ventilation during the sampling campaigns, since no new waste was stored in the facility during the period of this study.

From this study it was found that for four out of the total seven sampling campaigns the average ^{222}Rn concentration in the investigated radioactive waste storage facility exceeded the action level of 1000 Bq m^{-3} [9]. The correlation between work activity at the facility and average radon concentration showed that improvement in ventilation is one of the most effective radiation protection procedures to reduce radon exposure.

Table 2. Average radon concentrations and standard deviations in the radioactive waste storage facility (kBq m^{-3}).

Sampling point	Radon concentration (kBq m^{-3})						
	Jun–Aug 2012	Aug–Oct 2012	Oct–Nov 2012	Nov–Dec 2012	Dec–Jan 2012/2013	Jan–Feb 2013	Feb–Mar 2013
1	0.55 ± 0.05	0.66 ± 0.06	1.65 ± 0.06	0.87 ± 0.04	2.75 ± 0.07	4.39 ± 0.39	4.51 ± 0.40
2	0.77 ± 0.07	0.65 ± 0.03	1.66 ± 0.10	0.76 ± 0.09	2.65 ± 0.01	4.34 ± 0.05	4.52 ± 0.02
3	0.84 ± 0.04	0.75 ± 0.01	1.49 ± 0.2	0.83 ± 0.03	2.69 ± 0.14	4.58 ± 0.05	4.67 ± 0.07
4	0.79 ± 0.04	0.70 ± 0.04	1.49 ± 0.02	0.78 ± 0.06	2.66 ± 0.08	4.89 ± 0.08	4.24 ± 0.02
5	0.69 ± 0.01	0.70 ± 0.04	1.59 ± 0.06	0.84 ± 0.07	2.62 ± 0.05	4.76 ± 0.09	4.60 ± 0.27
6	0.69 ± 0.02	0.75 ± 0.01	1.52 ± 0.01	0.86 ± 0.10	2.75 ± 0.07	4.58 ± 0.23	4.79 ± 0.24
7	0.80 ± 0.02	0.65 ± 0.02	1.68 ± 0.01	0.82 ± 0.06	2.83 ± 0.02	4.58 ± 0.23	4.49 ± 0.04
8	0.67 ± 0.14	0.76 ± 0.06	1.74 ± 0.08	0.99 ± 0.03	2.7 ± 0.25	4.48 ± 0.21	4.39 ± 0.51
9	0.92 ± 0.08	0.89 ± 0.04	1.66 ± 0.13	0.83 ± 0.06	2.98 ± 0.21	4.65 ± 0.27	4.50 ± 0.24
10	1.14 ± 0.04	0.85 ± 0.06	1.95 ± 0.04	1.26 ± 0.04	2.92 ± 0.14	4.41 ± 0.65	4.95 ± 0.24
11	0.80 ± 0.08	0.68 ± 0.04	1.66 ± 0.02	0.81 ± 0.03	2.65 ± 0.09	4.47 ± 0.10	5.19 ± 0.45
12	0.73 ± 0.01	0.68 ± 0.01	1.50 ± 0.01	0.88 ± 0.06	2.65 ± 0.05	4.43 ± 0.18	4.52 ± 0.12
13[a]	0.31 ± 0.02	0.63 ± 0.01	ND[b]	0.69 ± 0.08	0.62 ± 0.03	0.73 ± 0.01	0.95 ± 0.06

[a] Antechamber.

[b] ND—not determined.

It should be added that unavoidably the radon concentration results took into account the radon from the soil under the construction. However, the radon exhalation rate from the soil around IPEN facilities was evaluated during the period from April 2012 to May 2013; the average value obtained of 0.026 Bq m^{-2} s^{-1} [20] is of the same order of magnitude of the estimated mean worldwide flux of 0.016 Bq m^{-2} s^{-1} from UNSCEAR [21]. Thus, based on the average radon exhalation rate from the soil around IPEN, and considering a theoretical very low ventilation rate and the facility dimensions, the radon concentration in the facility due to soil was estimated [21] to be about 250 Bq m^{-3}.

3.2. Effective dose due to radon inhalation

Inhalation is the main source of radon intake. The radiation risk is related to the effective dose received by an individual. This effective dose can be evaluated from dosimetric models or it can be inferred from the results of epidemiological studies. In this study, the effective dose due to ^{222}Rn inhalation was assessed considering the ICRP Publication 65 recommendations [7], the annual radon inhalation, an equilibrium factor of 0.4, an exposure time of 2000 h yr^{-1} and the effective dose conversion coefficient for ^{222}Rn. This dose conversion coefficient is based on the results of epidemiological studies, and it assumes that the aerosol conditions for the exposure were not too different from those for the uranium miner exposure.

Two average radon concentration scenarios were considered for dose assessment due to inhalation of radon at the investigated radioactive waste storage facility: a scenario with improved ventilation, when the facility remained open most of the time (August–October 2012), and another scenario, when the facility remained closed most of the time (December 2012 to March 2013). The effective doses varied from 4.2 mSv yr^{1} for the lower average radon concentration (August–October 2012) to 21 mSv yr^{-1} for the higher average concentration (December 2012 to March 2013). These estimations are quite conservative, since the working time hypothesis of 2000 h is rather moderate.

For the worst-case scenario, the results of the calculations show that the annual effective dose due to radon inhalation exceeds the occupational dose limit of 20 mSv yr^{-1} [22].

4. Conclusions

The present study aimed to contribute to the establishment of corrective actions, in order to reduce radon exposure for workers at the radioactive waste storage facility, located at the Nuclear and Energy Research Institute (IPEN/SP).

From the results obtained, it can be concluded that introducing a ventilation system could be an adequate corrective action to reduce exposure to within recommended levels.

It is important to point out that the national regulations establish the use of a ventilation system in interim storage facilities; the installation of such a system was contemplated in the original project and it will be installed in the near future.

However, it is necessary to know what the origin of the ^{222}Rn is, since the estimated radon concentration in the facility due to soil cannot be considered as significant. Hence new research will be carried out aiming at evaluating the radon exhalation from the different classes of radioactive waste. Only after this new research will it be possible to evaluate if there is some failure in the radioactive waste treatment processes and what kind of corrective actions could be taken to mitigate the radon exhalation.

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