

Radon in indoor concentrations and indoor concentrations of metal dust particles in museums and other public buildings

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Abstract The aim of this study was to evaluate the public and occupational exposure to radon and metal-bearing particles in museums and public buildings located in the city of Rio de Janeiro, Brazil. For this study, four buildings were selected: two historic buildings, which currently house an art gallery and

an art museum; and two modern buildings, a chapel and a club. Integrated radon concentration measurements were performed using passive radon detectors with solid state nuclear track detector-type Lexan used as nuclear track detector. Air samplers with a cyclone were used to collect the airborne particle samples that were analyzed by the particle-induced X-ray emission technique. The average unattached-radon concentrations in indoor air in the buildings were above 40 Bq/m³, with the exception of *Building D* as measured in 2009. The average radon concentrations in indoor air in the four buildings in 2009 were below the recommended reference level by World Health Organization (100 Bq/m³); however, in 2011, the average concentrations of radon in *Buildings A* and *C* were above this level, though lower than 300 Bq/m³. The average concentrations of unattached radon were lower than 148 Bq/m³ (4pCi/L), the USEPA level recommended to take action to reduce the concentrations of radon in indoor air. The unattached-radon average concentrations were also lower than the value recommended by the European Union for new houses. As the unattached-radon concentrations were below the international level recommended to take action to reduce the radon concentration in air, it was concluded that during the period of sampling, there was low risk to human health due to the inhalation of unattached radon in these four buildings.

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Introduction

Human exposure occurs when a person comes into contact with a pollutant of a certain concentration during a certain period of time (Ott et al. 2007). Based on this definition, people who live in big cities are continually exposed to different sources of pollution. The main pathways of intake of a compound and/or an element are inhalation and ingestion, although dermal contact can be also considered as a pathway. The majority of studies for the risk assessment due to human exposure to toxicity substances only consider outdoor sources of pollution; however, indoor aerosols can also cause serious damage to human health (IARC 1988; UNSCEAR 2000; NCI 2007; Stranger et al. 2007). Indoor aerosols are generated inside microenvironments that are primarily closed rooms with heating and/or cooling systems and improper exhaust ventilation. Currently, people remain long periods of time inside these closed rooms working, studying or otherwise, increasing exposure to aerosol particles present in the indoor air (Lee et al. 2001, 2002a, b; Li et al. 2001). From this chronic exposure to indoor pollutants, the concept of the sick building syndrome (SBS) emerges, a commonly used term for symptoms resulting from problems related to poor indoor air quality (IAQ). Complaints common to SBS include allergic rhinitis, headaches, flu-like symptoms, watering of the eyes and difficulty breathing (Mishra et al. 1992). Radon concentrations and metal-bearing particle concentrations are parameters that should be considered to evaluate the IAQ.

The aim of this study was to evaluate the public and occupational exposure to radon and metal-bearing particles in museums and public buildings located in the city of Rio de Janeiro, Brazil. For this study, four buildings were selected: two historic buildings, labeled *Buildings A* and *C*, which currently house an art gallery and an art museum, respectively, and two modern buildings, labeled *B* and *D*, housing a chapel and a club, respectively.

Radon (Rn) is a radioactive noble gas; the ^{222}Rn and ^{220}Rn are two radon isotopes, and they are the decay products of ^{238}U and ^{232}Th , respectively. ^{222}Rn and ^{220}Rn decay emitting an alpha particle with half-lives of 3.8 days and 55.6 s, respectively. ^{238}U and ^{232}Th are widely spread in soil and rocks throughout the earth's crust. They are present in trace concentrations in granites and in minerals such as phosphate ores that are

used to produce gypsum, etc., and from which radon gas can emanate. Granite and gypsum board are widely used building materials, which can thus affect IAQ. The radon gas particles can be present in the air as unattached-radon particles and/or as attached-radon particles. Both types of radon particles can be inhaled and the radon decay products, which are solid particles, decay emitting alpha or beta particles that are then deposited in the lungs. Radon exposure is considered the second leading risk factor of lung cancer after tobacco smoking, studies show that there is a correlation between radon exposure and lung cancer occurrence. Radon is an element classified as carcinogenic class I by the IARC—International Agency for Research on Cancer (IARC 1988; UNSCEAR 2000). The estimated value of worldwide average annual dose to the public due to natural exposure is about 2.4 mSv, and approximately 50 % of this dose is due to radon exposure (UNSCEAR 2000, 2008).

During the last decades, some studies concentrated on determining the risk assessment due to indoor radon exposure, including in dwellings and workplaces (Swedgemark and Mjönes, 1984; EPA 1991; Tsai et al. 1995; Giovani et al. 2001; Katja Radon et al. 2002; Orlando et al. 2002; Magalhães et al. 2003; Vincent and Werner 2003; Bochicchio et al. 2005; Carelli et al. 2009; Klinmalee et al. 2009; Khan and Puranik 2011; López-Coto et al. 2012; Trevisi et al. 2012). The results of these studies showed that the radon concentrations vary widely from place to place.

Moreover, in closed rooms, people are exposed to metal-bearing particles. These particles can be generated by indoor sources such as cleaning products and smoke from fireplaces and cigarettes; however, these particles can also be generated outside the buildings and be transported by the ventilation system and/or through the windows from outside area into the buildings (EPA 1991; Pegas et al. 2011).

Art galleries and museums are usually closed rooms with heating and/or cooling systems, with controlled temperature and humidity, resulting in a low renewal rate of the air. These areas are visited daily by hundreds of visitors who are exposed to metal-bearing particles and to radon and its decay products. Moreover, the workers in museums and in art galleries usually work for long periods of time in these closed rooms, and consequently, they are occupationally exposed to metal-bearing particles and to the radon and its decay products.

There are many factors to air quality in Rio de Janeiro, including pollution, coastal air and weather. Rio de Janeiro is the second largest city in Brazil, boasting approximately 10 million people within the city proper, making it the 6th largest in the Americas and 26th in the world. The city of Rio de Janeiro is within the state of Rio de Janeiro, and only the city is addressed in this work. Rio de Janeiro has a tropical climate that closely borders a tropical monsoon climate, and is often characterized by long periods of heavy rain from December to March. The city is along the Atlantic Ocean coast. Inland temperatures above 40 °C are common during the summer, though rarely for long periods, while maximum temperatures above 27 °C can occur year round. There are more than 15 museums and 50 art galleries in Rio de Janeiro, which are visited by hundreds of people, mainly children and seniors. Due to the weather conditions, the majority of museums, art galleries and public buildings are closed rooms with air condition system. However, there are no data on the air quality inside museums, art galleries and public buildings in Rio de Janeiro.

Methods

Radon samples were collected in four buildings located in Rio de Janeiro city in 2009 and 2011. The buildings were identified as *Building A*, *Building B*, *Building C* and *Building D*. The aerosol samples only were collected in two buildings: *Buildings A* and *C*.

Characteristics of the buildings

Building A is an art gallery; it was built in 1826 using stone, granite, marble, clay, sand, whale oil and wood as building materials. This building was designed as a residence with large windows and doors. The building was designed with three floors; the first floor was an open area, with columns that support the second and the third floors. The second and third floors were used as a residence and the materials used to cover the floor are marble, granite and wood, and the walls were painted using lime. In 1994, the second and third floors were restored to house an art gallery, and they remain as they were originally designed in 1826. Approximately 5 years ago, the first floor was remodeled to house an office, and the open areas between the columns were closed using glass, bricks and cement.

Building B is a modern chapel; it was built in 2005 using bricks, cement, concrete, marble and granite. The chapel has two large doors that remain open during the day.

Both *Building A* and *Building B* are in Gávea, a neighborhood located in the south zone of Rio de Janeiro, Brazil. Gávea sits right at the edge of the Tijuca Forest (a national forest), and tree-covered mountains make up the north of the area.

Building C is an art museum; it was originally built in 1603 using stone, clay, sand, whale oil, granite, marble and wood. In subsequent years, the building was remodeled several times to increase the number of rooms. In 1922, it was restored to house an art museum.

Building D is a club; it was built in 1946 using bricks, cement and concrete.

Buildings C and *D* are located in downtown; they are surrounded by streets with intense traffic, and both buildings are located 2 km from the regional airport and 10 m from Guanabara Bay.

Radon sampling

The radon samples were collected inside the four buildings in 2009 and in 2011; the number of samples per building is shown in Table 1.

The radon concentration measurements were taken using passive radon detectors. Each passive radon detector contains a 35-mm-diameter polypropylene chamber with a glass fiber filter used to retain solid particles. Solid state nuclear track detectors (SSNTDs) were located behind the filter. Lexan was used as nuclear track detector. In order to determine the number of tracks in a Lexan detector exposed to radon, each detector was chemically pre-etched (1,000 V at 100 Hz) for 40 min and then was etched (800 V at 3,000 Hz) at 30 °C in a solution of KOH (6 N) and C₂H₅OH for 3 h. The pre-etching timing was chosen to obtain alpha particle tracks of about 100 μm, which could be easily identified and visualized by computer scanning within 1,200-dpi resolution. The Lexan detectors were scanned, and the number of tracks per square centimeter was determined using specially developed software (Pinheiro 2009). More details concerning the methodology and calibration have been described elsewhere (Baixeras et al. 1997; Möre and Hubbard 1997; Pressyanov et al. 2004; Urban and Piesch 1981; Magalhães et al. 2003). The number of tracks observed in the detector was related to radon

Table 1 Average of indoor radon concentrations and the standard deviations calculated for each building

Building	Years	Number of samples	Concentrations (Bq/m ³)			
			Average	Standard deviation	Minimum	Maximum
A	2009	26	58	27	14	158
	2011	25	146	42	87	247
B	2009	9	48	13	32	67
	2011	10	136	26	85	166
C	2009	21	87	27	47	136
	2011	21	95.2	26	60	150
D	2009	26	36	9.1	22	55
	2011	23	70	19	41	122

concentrations in the air using a determined calibration factor. During each sampling four detectors were kept for 3 months inside a closed bag in the laboratory. These detectors were analyzed with the detectors used to collect the unattached-radon samples.

Air samples

Air samplers with a cyclone were used to collect airborne particles samples inside *Buildings A* and *C*. The sampling sites were the same closed rooms selected to collect the radon samples; however, the air samplers were located in the middle of the rooms while the radon detectors were fixed 5 cm from the walls. The samples of airborne particles were collected during 5 consecutive days within the period of radon sampling. The fine fraction of aerosol (particles with aerodynamic diameter <2.5 μm) was collected on a Nuclepore filter, and these filters were fixed in a sample support disk for particle-induced X-ray emission (PIXE) analysis without any previous preparation.

PIXE technique

The air samples were analyzed by particle-induced X-ray emission (PIXE) technique to determine the concentrations of elements in the fine fraction of aerosol. This analysis was performed using a 2-MeV proton beam obtained from a 4.0-MV van de Graaff accelerator with a target current of ~20 nA at the van de Graaff Laboratory at the Pontifical Catholic University of Rio de Janeiro (PUC-Rio). Further details of this experimental arrangement are described in the literature (Dias da Cunha et al. 2000). The X-rays emitted from the samples were detected using a Si-PIN

detector with a 0.2-μm-thick aluminum absorber foil. The X-ray spectra were analyzed using custom-designed software based on the stripping of a multi-element spectrum.

Results

Unattached-radon concentrations

The average of indoor unattached-radon concentrations and the standard deviations, calculated for each building, are presented in Table 1. Radon values are expressed in terms of average activity concentrations (Bq/m³).

The average radon concentrations in indoor air in 2009 were lower than in 2011, but in both samples, the average radon concentrations were higher in the historic buildings *A* and *C* than in the modern buildings *B* and *D*. The concentrations in the historic buildings and in the modern ones varied from 14 to 247 Bq/m³ and from 22 to 166 Bq/m³, respectively. However, the average values were below the average radon concentrations determined by Travesi et al. (2012) at workplaces in Italy (153 Bq/m³). The average radon concentrations in indoor air in the buildings were also above the average radon concentrations in residences located in Rio de Janeiro (40 Bq/m³) (Magalhães et al. 2003), except for the low average concentration in *Building D* in 2009. The World Health Organization (WHO) in 2010 presented a recommended reference level (the national reference level) of 100 Bq/m³ for radon in dwellings. The recommendation also says that where this is not possible, 300 Bq/m³ should be selected as the highest level. The average concentrations of radon in indoor air

in the 2009 data were below the recommended reference level by WHO (100 Bq/m³); however, in 2011 the average concentrations of radon in *Buildings A* and *B* were above this level, though lower than 300 Bq/m³. The average radon concentrations in the buildings in 2009 and 2011 were all lower than 148 Bq/m³ (4pCi/L), the level at which the United States Environmental Protection Agency (USEPA) recommends taking action to reduce the concentrations of radon in indoor air (USEPA 2007). The average radon concentrations in the buildings were also below the value recommended by the European Union for new houses (200 Bq/m³) (EUR 1995). Although the average radon concentration in indoor air in residences located in Rio de Janeiro was lower than the average radon concentrations in the buildings in this study (except *Building D* in 2009), these values were lower than the international recommended levels to take an action to reduce the concentration.

The distributions of the radon concentrations in each building were compared using a nonparametric statistical Mann–Whitney rank test, with a 95 % confidence level (Zar 1999).

These results showed that the distribution of radon concentrations in all buildings varied during the sampling, and the unattached-radon concentrations in air samples collected in 2009 were lower than in samples collected in 2011. In 2009, the temperature inside the buildings varied from 25.7 to 26.6 °C and relative

humidity varied from 83 to 84 %. In 2011, the temperature inside the buildings varied from 25.3 to 28 °C and the relative humidity varied from 74 to 77 %. The rainfall index in Rio de Janeiro during sampling periods was 900 mm in 2009 and 101 mm in 2011. The passive detector assembly has a filter to prevent particles with diameters larger than diameters of the unattached-radon particles from reaching the Lexan detector. The radon tends to attach to water molecules, so the diameters of attached-radon particles are larger than the diameters of unattached-radon particles, and so due to the higher precipitation in 2009 than in 2011, the unattached-radon concentrations in indoor air were reduced. The statistical analyses also show that the distributions of radon in the buildings were not equal and suggest that the rainfall index caused a variation in the unattached-radon concentrations in indoor air.

Metal-bearing particle concentrations

In order to evaluate the transport of airborne particles from the outside to the inside of the buildings, concentrations of metal-bearing particles in the fine fraction of aerosols were determined inside *Buildings A* and *C*. The results were compared to the metal concentrations in the surrounding areas of the buildings, which were available in the literature. The metal concentration values are presented in Table 2.

Table 2 Metal concentrations in indoor air in *Buildings A* and *C*

Element	Concentrations (µg/m ³)							
	Average		Standard deviation		Minimum		Maximum	
	A	C	A	C	A	C	A	C
S	<DL	4.6E+00	<DL	3.8E+00	<DL	7.0E+01	<DL	1.8E+02
Cl	1.2E+02	2.5E-01	7.6E+01	1.9E-01	7.0E+01	9.2E-04	1.8E+02	1.6E+00
K	7.6E-01	<DL	8.1E-01	–	9.2E-04	<DL	1.6E+00	<DL
Ca	2.9E-01	<DL	5.4E-01	–	2.1E-01	<DL	1.2E+00	<DL
Cr	1.5E-03	9.9E-04	9.1E-04	6.9E-04	5.2E-04	1.0E-02	3.2E-03	2.1E-02
Ti	1.6E-02	<DL	7.7E-03	–	1.0E-02	<DL	2.1E-02	<DL
Mn	2.4E-04	1.8E-04	1.7E-04	2.1E-04	7.6E-05	1.2E-04	6.8E-04	2.5E-04
Fe	1.4E-02	5.0E-03	2.4E-02	6.5E-03	1.3E-03	4.0E-02	9.1E-02	9.1E-02
Ni	3.3E-04	3.5E-04	2.0E-04	2.5E-04	4.0E-05	4.0E-05	6.3E-04	6.0E-05
Cu	6.6E-04	1.2E-04	8.1E-04	2.5E-05	7.5E-05	1.0E-03	2.3E-03	2.3E-03
Zn	8.5E-04	3.6E-04	8.3E-04	2.1E-04	1.9E-04	1.7E-03	2.4E-03	2.4E-03

<DL is below the detection limit

S DL = 2.7 ng/m³; K DL = 2.7 ng/m³; Ca DL = 0.08 ng/m³; Ti DL = 0.02 ng/m³

Particles containing Cl, K, Ca, Cr, Ti, Mn, Fe, Ni, Cu and Zn were identified in the aerosol samples collected in *Building A*. These results were compared to results from previous studies in the area surrounding *Building A* (Lizarraga et al. 1995; Dias da Cunha et al. 2000).

These previous studies identified chlorine-bearing particles, sodium-bearing particles and sodium chlorite particles, which characterize marine aerosols in the area surrounding the building. These particles were also identified in indoor air, suggesting that there was a contribution of airborne particles from the outside to the inside of the building. K-, Ca-, Ti- and Mn-bearing particles could have been generated by building materials worn away by use or wind, but these elements are also present in soil dust particles, and they were identified in the previous studies to characterize the air quality in the area surrounding the building. Moreover, K-bearing particles and Ca-bearing particles could be generated by the vegetation that surrounds *Building A*. The airborne particles containing Cr, Ni and Cu were identified in indoor air but were probably generated outside the building, because possible sources of these elements were not identified inside the building. The main sources of these elements are the anthropogenic sources such as mechanical engine wear, and the burning of the fossil fuel also generates the Ni-bearing particles. These particles were also identified in previous studies in Gávea (Lizarraga et al. 1995; Dias da Cunha et al. 2000). Fe-bearing particles could be generated by the wearing down of doorknobs and hinges, which were manufactured using iron. However, Fe-bearing particles and Zn-bearing particles could be generated by the vegetation surrounding the building and/or anthropogenic source such as burning of fossil fuel, as mentioned before. These particles were also identified in previous studies (Lizarraga et al. 1995; Dias da Cunha et al. 2000).

S, Cl, Cr, Mn, Fe and Ni were identified in the indoor air in *Building C*. S-bearing particles and Cl-bearing particles indicate the presence of marine aerosol inside the closed rooms in *Building C*. However, S-bearing particles can be generated by anthropogenic sources, since S is present in the fossil fuel used by cars and in fuel used in the commercial airplanes. Particles containing Cr and Ni also characterize anthropogenic sources of aerosols, because Cr and Ni are also present in the fossil fuel and in oils

used to lubricate car engines. These particles indicate that the main anthropogenic sources were the intense car and bus traffic using fossil fuel, and/or the local airport located 2 km from *Building C*. Nevertheless, K, Ca and Ti were not identified in indoor air in *Building C*, suggesting that there were no sources of these particles, either natural or anthropogenic. These results showed that the air quality in the environment outside the building could be a source of aerosols inside the building.

Conclusions

The unattached-radon concentrations in indoor air in the four buildings varied; in 2009, the unattached-radon concentrations were lower than in 2011. These variations in unattached-radon concentrations could be caused by weather conditions, as discussed above. The rainfall index during the samples collected in 2009 was 900 mm, while in 2011 it was 101 mm, which could have reduced the unattached-radon concentrations in indoor air in the buildings in 2009. The average unattached-radon concentrations in indoor air in the buildings were above 40 Bq/m³, except the average concentration in *Building D* in 2009. In 2009, the average unattached-radon concentrations in indoor air in the four buildings were below the recommended reference level by WHO (100 Bq/m³); however, in 2011, the average concentrations of unattached-radon in buildings *A* and *B* were above this level, though lower than 300 Bq/m³. The average concentrations of unattached-radon were also lower than 148 Bq/m³ (4pCi/L), the USEPA level recommended to take an action to reduce the concentrations of radon in indoor air, and lower than the value recommended by the European Union for new houses. As the unattached-radon concentrations were below the international level recommended to take an action to reduce the radon concentration in air, it was concluded that during the period of samples, there was no heightened risk to human health due to the inhalation of unattached-radon in these four buildings.

Particles that characterize marine aerosols and anthropogenic sources were identified inside the buildings. Based on these results, it was concluded that there was a possible contribution of particles from outside sources to the indoor air although these buildings were closed buildings.

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