Original Article

LEVELS OF NATURAL RADON-RADIOACTIVITY IN THE SÃO VICENTE, SP, ROCK MASSIF*

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Abstract

OBJECTIVE: The objective of this study was to perform a passive and time-integrated radon monitoring in several soil and water samples and indoor environments of the São Vicente, SP, rock massif with the purpose of evaluating the presence and distribution of that radioactive gas in this region. MATERIALS AND METHODS: The technique employed consisted of exposing Makrofol E-type polycarbonate plastic detectors (SSNTD – solid-state nuclear track detectors), using the closed cup method, to radon emanated from ground water samples and to the gas accumulated inside indoor environments (dwellings) and inside rock cracks existing in the São Vicente rock massif. RESULTS: The radon concentration values obtained ranged from 8.1 to 36 Bq/l in natural groundwaters; between 68 and 610 Bq/m³ in dwellings; from 0.41 to 3.46 kBq/m³ in soils and from 0.72 to 5.85 kBq/m³ inside rock cavities of the São Vicente rock massif. CONCLUSION: In some dwellings and in most of groundwater samples the radon concentration values found in this study

have exceeded the maximum levels proposed by international agencies. Thus, intervention actions are recommended for radon dissipation in dwellings and during consumption water collection.
Keywords: Radon; Natural radioactivity; Nuclear track detectors; Environment; Uranium.

INTRODUCTION

Radon is a natural inert gas formed by disintegration of radium, an element of uranium and thorium decay series. It is found in practically every place, emanating naturally from the Earth crust, and for being a gas, has the property of accumulating in indoor environments like dwellings, buildings, caves, mines and tunnels. The ^{222}Rn isotope is an alpha emitter ($T_{1/2}$ = 3.82 days, E_{α} = 5.49 MeV) and, together with its non-gaseous daughter products, ^{218}Po and ^{214}Po, accounts for approximately 50% of the effective dose equivalent produced by natural ionizing radiation[(1,2)]. The ingestion of water as well as inhalation of air containing high levels of this gas, may represent a direct risk to the population health, since sensitive cells in the respiratory and gastrointestinal tracts are exposed to ionizing radiations, leading to illness due to the occurrence of some types of cancer in these organs[(3,4)]. This fact has been demonstrated by epidemiologic studies developed with mining workers in several countries[(5)], and presently the radon is already classified by the International Agency for Research on Cancer (IARC) as a class I carcinogen[(6)].

In recent measurements of radon performed in different types of natural water in the Baixada Santista region[(7)], values higher than the 11.11 Bq/l maximum level of contamination (MLC) recommended by the United States Environmental Protection Agency (USEPA) have been observed[(8)], in samples from two water sources localized in the São Vicente mountain range (São Vicente Rock Massif). The Brazilian regulation (Health Ministry Decree 1469 dated 12/29/2000) does not mention specifically the radon gas and establishes a maximum value permitted for total alpha-activity in consumption water – 0.1 Bq/l – that is, it considers only non-gaseous alpha emitters as radioactive contaminants. This rock complex is between the cities of Santos e São Vicente and is formed by a range of hills with at maximum 220 m in height, presenting high grade of metamorphism with predominant gneissic and granitic rocks as well as post-tectonic granitoid formations of precambrian age[(9)]. These hills are densely populated, principally on hillsides, and have sources of natural waters which are much utilized for consumption by the local population. Therefore, determining the radon ratio in several types of environmental samples collected in this rock complex is important for general radiological protection of the population.

The objective of this study was an integrated and passive monitoring of the radon ratio in samples of groundwaters, dwellings, fissures and soil from the São Vicente rock massif, by means of the SSNTD technique (solid state nuclear track detector), aiming at evaluating the level of exposure to which the local population is being submitted.

MATERIALS AND METHODS

Figure 1 shows the locations selected for groundwaters collection and local direct measurements of radon gas in dwellings, fissures or cracks existent in rocks, as well as in the soil of the São Vicente rock massif.

The technique of alpha particles tracks recording in SSNTD-type detectors was employed for radon integrated and passive monitoring[7]. The technique consisted of exposing Makrofol E-type polycarbonate plastic films (200 μm thickness), in a fixed-geometry device, to radon released from water samples (Figure 2) during a period of approximately 30 days. As the ^{222}Rn half-life is of 3.82 days, one has estimated that, in this time interval, 99.5% of radon nuclei contained in these samples would have decayed. In the case of continuous detection of radon in soil, the device has been positioned at a 15 cm depth (Figure 3), remaining in place for a period ranging between two and three months. A control exposure was performed in the city o Guarujá, in lowlands sandy soil, therefore a very different type from that studied.

For monitoring radon in dwellings and fissures or cracks in rocks, one has fixed the detector device directly in the places selected. The exposure-to-radon period in these places was about three months. In the detector, the alpha-particle derived from the radon radioactive decay as well as its daughter products remain recorded on the plastic film in the form of trails or pits. After irradiation or exposure, the plastic films were removed from the detectors and developed in an appropriate PEW solution 45% water, 40% ethylic alcohol and 15% KOH) at 70ºC for 120 minutes, so that the trails and pits could be made visible under an optical microscope[7]. Afterwards, the trails and pits were counted by means of a computer (PC) coupled with a video camera and an ordinary optical microscope, with a resulting 620 times image enlargement. The detector device response or efficiency function was determined with a calibrated Pylon RN 150-3 model radon chamber existent in the Environmental Division of IPEN-CNEN/SP. This chamber includes a ^{226}Ra source and has been standardized by its manufacturer aiming at releasing ^{222}Rn gas activity (2.48 ± 4%) Bq to the system to be calibrated. The detector device, after being removed from the camera, remained closed for a week for following-up of the decay of radon stored inside it, by means of evaluation of alpha particles trails and pits on the Makrofol E plastic film.

RESULTS

For detector response function determination, four exposures in the Pylon chamber were performed and the mean value obtained was (0.158 ± 0.007) trails.cm^{-2}.d^{-1}/Bq.m^{-3}.

Table 1 shows the values obtained in this study for mean levels of radon in samples collected from natural sources of water (F) in the São Vicente rock massif and Table 2 shows concentrations of this gas (R) in dwelling localized in this region. Results from measurements of radon levels in this complex rock cavities soil (S) and air (A) can be seen on Table 3. The total uncertainty of the measurements has been calculated taking the following sources of partial errors into consideration: statistical errors (1.53% to 22%), detector calibration (4.7%) and discount of the background radiation (7.4%).

As it may be observed in Table 1, except for the Marapé source, all the water samples analyzed in this study have presented higher radon concentrations than the maximum contamination level recommended by USEPA (11.11 Bq/l)[8]. However, it is important to note that the radon level found in each sample corresponds to the value obtained directly at the respective source and certainly should not be the final concentration of this gas at the moment of consumption by the population due to losses during bottling, transportation and handling. This has been experimentally proved by means of a simulation of collection and transportation of water from the São Jorge source. The collection and transportation of this water to the laboratory was made in a five-liter glass bottle. Afterwards, measurements of radon levels in one-liter samples withdrawn from that bottle were performed at time intervals of about one hour. Early in the first sampling there was a remarkable decrease of one factor 4 in the final radon activity.

In Table 4 the results obtained in this study are compared with those published in the literature and by other authors in similar studies, taking geological differences into consideration. Thus, provided geological differences between places studied are considered, one may observe that there is reasonable concordance among authors.

DISCUSSION

According to the International Commission for Radiological Protection (ICRP)[1], in the case of radon-in-air inside dwellings, the activity level recommended for intervention is between 200 and 600 Bq/m^3, provided a 7,000 hours/year occupation time by population is considered. This study results show that in two residences (Nova Cintra and Voturuá) the limit is being surpassed, therefore, it is recommended that interventional actions are adopted for radon levels reduction in these places.

As regards radon levels found in São Vicente rock massif soils, the values were constantly much higher than the value obtained for the control place, therefore indicating the presence of relatively higher track quantities of uranium, thorium and radium ores in this rock complex.

Radon concentrations usually found in these types of samples and environments are in direct connection with the geological formation of the region, since the types of rocks forming these soils may present higher or lower concentrations of uranium, thorium and radium ores which are natural generators of radon gas. This gas may diffuse through rock fissures or being transported from the subsoil to the surface by means of water flows so constituting an exposure source for the population living of visiting such places.

The present study results regarding radon concentrations in water, dwellings, soil, fissures and caves localized on the São Vicente mountain range prove a relatively high occurrence of this gas, in some cases reaching higher values than the internationally recommended limits. Therefore, it is important to perform periodical monitoring of the radon gas levels inside dwellings localized near the rock complex to know if they are in compliance with international recommendations.

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NÍVEIS DE RADIOATIVIDADE NATURAL DECORRENTE DO RADÔNIO

Figuras e Tabelas

Figure 1. Illustrative map of São Vicente rock massif indicating places selected for water samples collection (F) and direct radon gas monitoring in dwellings (R), soils (S) and rock cavities (A).

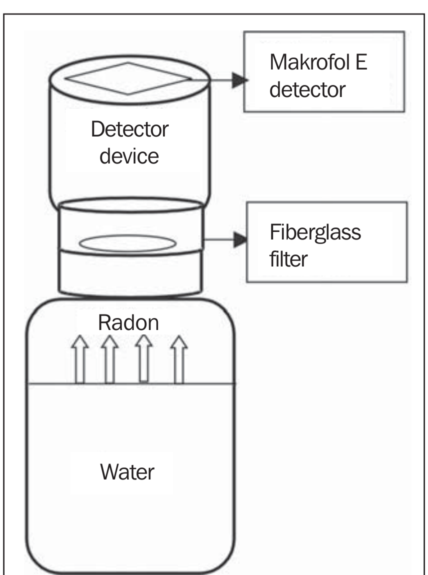

Figure 2. Diagram of the experimental arrangement utilized for radon passive and integrated monitoring of water samples collected in the São Vicente mountain range.

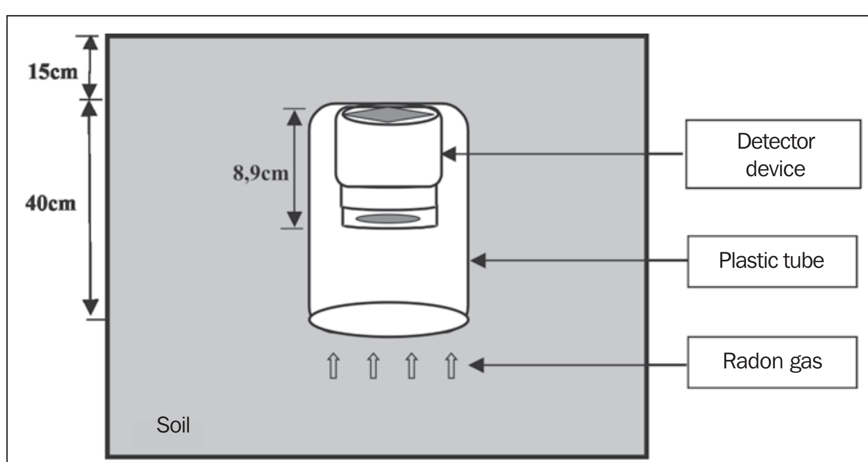

Figure 3. Diagram of the experimental arrangement utilized for radon passive and integrated monitoring in the soil of the São Vicente mountain range.

Table 1 Average radon rate in natural water sources of the São Vicente mountain range rock complex.

Sources	Location	Radon concentration (Bq/l)
Biquinha de Anchieta	F1	32.2 ± 7.4
São Jorge	F2	36.0 ± 9.5
Nova Cintra	F3	13.2 ± 1.3
Marapé	F4	8.1 ± 1.0
Caneleira	F5	19.5 ± 1.7
Jabaquara	F6	18.7 ± 1.7

Table 2 Radon concentration in dwellings localized on São Vicente mountain range rock complex.

Dwellings	Location	Radon concentration (Bq/m^3)
Monte Serrat	R1	163 ± 18
São Bento	R2	118 ± 14
Jabaquara	R3	112 ± 11
Nova Cintra	R4	610 ± 55
Nova Cintra	R5	68 ± 7
Voturuá	R6	145 ± 14
Voturuá	R7	307 ± 63

Table 3 Radon levels in soil and fissures of the São Vicente mountain range rock complex.

Samples	Location	Radon concentration (Bq/m^3)
Solo – Voturuá	S1	865 ± 80
Solo – Voturuá	S2	2,262 ± 208
Solo – Voturuá	S3	1,600 ± 149
Solo – Nova Cintra	S4	3,462 ± 320
Solo – Nova Cintra	S5	996 ± 93
Solo – Controle	S6	413 ± 45
Cavidade – Voturuá	A1	1,683 ± 370
Cavidade – Nova Cintra	A2	5,852 ± 527
Cavidade – Nova Cintra	A3	719 ± 85

Table 4 Comparison of radon concentration intervals found in different natural water sources, dwellings, soils and caves or fissures.

Types of samples	Radon concentration	Reference	Localization
Natural water sources	24 to 40 Bq/l	Hopke *et al*.[10]	United States
Natural water sources	0.9 to 35.4 Bq/l	Alabdula'aly[11]	Saudi Arabia
Natural water sources	0.1 to 576 Bq/l	Horváth *et al*.[12]	Venezuela
Natural water sources	5.3 to 83.7 Bq/l	Silva *et al*.[13]	Brazil (Recife)
Natural water sources	8.1 to 36 Bq/l	This study	Brazil (Santos)
Dwellings	14.3 to 135 Bq/m^3	Canoba *et al*.[14]	Latin America
Dwellings	95 to 202 Bq/m^3	Singh *et al*.[15]	India
Dwellings	< 5 to 200 Bq/m^3	Magalhães *et al*.[2]	Brazil (Rio Janeiro)
Dwellings	56 to 168 Bq/m^3	Geraldo *et al*.[16]	Brazil (Baixada Santista)
Dwellings	68 to 610 Bq/m^3	This study	Brazil (São Vicente rock massif)
Soils (0.5 m)	0.8 to 26.7 kBq/m^3	Abumurad[1]	Jordan
Soils (0.08 m)	1.89 to 17.95 kBq/m^3	Choubey *et al*.[17]	India
Soils (1 m)	2.5 to 47.5 kBq/m^3	Jönsson[18]	Sweden
Soils (0.15 m)	0.41 to 3.46 kBq/m^3	This study	Brazil
Caves in rocks	0.07 to 12.55 kBq/m^3	Gillmore *et al*.[19]	United Kingdom
Caves in rocks	0.18 to 7.12 kBq/m^3	Lario *et al*.[20]	Spain
Caves in rocks	< 0,02 to 9,25 kBq/m^3	Solomon *et al*.[21]	Australia
Caves in rocks	0.10 to 80 kBq/m^3	Sajó-Bohus *et al*.[22]	Venezuela
Caves in rocks	0.72 to 5.85 kBq/m^3	This study	Brazil