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EXPOSURE ASSESSMENT DUE TO BUILDING MATERIALS IN ORDINARY HOUSES AT SÃO PAULO, BRAZIL

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ABSTRACT

The total effective dose rate to dwellers of ordinary houses at São Paulo, Brazil was obtained as a result of both external exposure from gamma emitting radionuclides in the building materials and internal exposure from inhalation of radon and their decay products. Samples of Brazilian raw materials and building products were analyzed by high resolution gamma spectrometry for their ^{226}*Ra,* ^{232}*Th and* ^{40}*K concentrations. Ambient levels of radon indoor were measured with solid state track detectors. The values obtained for effective dose rate are underneath the 2.4* $mSv \cdot y^{-1}$ *UNSCEAR assumed value for natural radiation sources.*

Keywords: building materials, gamma-ray spectrometry, radon

RESUMO

A taxa de dose efetiva total recebida por moradores de casas populares em São Paulo, Brasil foi obtida como resultado da exposição externa devido à presença de radionuclídeos emissores gama nos materiais de construção e da exposição interna devido à inalação de radônio e seus produtos de decaimento. Amostras de materiais de contrução brasileiros foram analisadas por espectrometria gama de alta resolução para a determinação das concentrações de ^{226}*Ra,* ^{232}*Th e* ^{40}*K. Níveis de radônio no interior das moradias foram medidos com detetores sólidos de traços nucleares. Os valores obtidos para a taxa total de dose efetiva estão abaixo do valor de 2,4* $mSv \cdot a^{-1}$*, assumido pela UNSCEAR, para exposição às fontes naturais de radiação.*

Descritores: mateirais de contrução, espectrometria gama, radônio

INTRODUCTION

Mankind's interest in the level of natural radiation exposure has strongly increased over the past fifty years, as people became aware that exposures from the use of nuclear technology for medicine, energy and weapons are far below those from natural sources, which represent approximately 70% of the radiation exposure received by the general population [1], [2].

Building materials are one of the most important sources of radiation exposure in dwellings, mainly because the gamma rays emitted by the long-lived ^{40}K and members of the ^{238}U and ^{232}Th chain. According to UNSCEAR [2], 11% of the effective dose from natural radiation received by population is caused by radon entry from building materials. So, it is very important to assess the exposure due to building materials, since most individuals spend 80% of their time indoors.

Natural radionuclides in building materials and the radon indoor concentrations have been investigated by several authors [3], [4], [5], [6], [7]. However, even knowing that some sites in Brazil present igneous rocks and abnormally high concentrations of radioactive minerals in the soil [8], there are few information [9], [10], [11], [12] about Brazilian raw materials and building products derived from those soils and rocks.

Since building material is a potential source of elevated radiation exposure and particularly concrete, which contains elevated levels of natural radionuclides, seems to be the strongest radon emanator, it is important to assess the exposure levels in houses constructed mainly of concrete. The present study was realized in a condominium having one hundred of popular houses built of concrete blocks at Santo André district of São Paulo, Brazil.

This paper presents the results of gamma ray spectrometry measurements of the naturally occurring radionuclides concentrations in the building materials utilized and the radon concentration in the dwellings. The internal and external dose rates for the people living in these houses were assessed using the UNSCEAR [1] procedures.

MATERIALS AND METHODS

All houses were built with the same materials, in the same manner, an unique 15 m^2 bedroom and bathroom, with only one door and one window. The walls are made of concrete, the floor of cement and the roof is a concrete beam. The materials used in the construction were: sand, cement, broken stone, concrete, roofing brick and concrete beam, all of same origin and were supplied by the Santo André municipallity according to their availability.

A total of 38 samples were prepared and analyzed for the two raw materials (sand and broken stone) and four building products (cement, concrete, roofing brick and concrete beam). The number of samples of each material is described in Table 1.

Solid samples as concrete beam, roofing concrete bricks and concrete blocks were crushed into about 10 mm particles before being sealed in 860 mL Marinelli polyethylene beakers for a 4 weeks ingrowth period. The cement, sand and broken stones samples were directly sealed in the Marinelli beakers.

The activity concentrations measurements were performed by using a high resolution 15cm^3 HPGe detector (EG&G, Ortec, USA), coupled to a 4K memory Ortec 918-A ADCAM multichannel buffer and 476-8 multiplexer and to a 386 PC/AT computer. The effective measured energy resolution for the 1.33 Mev ^{60}Co gamma transition is 1.7 keV.

The background distribution was obtained by measuring de-ionized water in the same sample geometry of 860 mL Marinelli polyethylene beaker.

All spectra were analyzed with the MicroSAMPO software for personal computer analysis of gamma-ray spectra from HPGe detectors [13]. The self-absorption correction of the samples was achieved using a pre-determined standard efficiency curve [14].

Indoor radon concentrations were determined by passive detection method with solid state nuclear track detectors using Makrofol E inside a diffusion chamber. A fiber-glass filter inside the chamber was used in order to stop the aerosols and radon daughters entering the chamber while allowing the radon to pass through it. Two diffusion chambers were used for each house and exposed during three months. The while surveillance covered the four seasons of the year.

RESULTS AND DISCUSSION

Radionuclide Concentrations. For the evaluation of the external gamma-ray indoor exposure it were determined the ^{226}Ra, ^{232}Th and ^{40}K concentrations in the building materials.

The ^{226}Ra concentration was determined assuming radioactive equilibrium in the ^{238}U chain, through the gamma transitions of 352 keV from ^{214}Pb and 609 keV from ^{214}Bi. Radium concentration for each sample was calculated as the mean value of those gamma transitions. The ^{226}Ra medium concentration for all samples of the same material was calculated as the mean value of the radium concentration of each sample.

The ^{232}Th concentration was determined assuming radioactive equilibrium in the ^{232}Th chain, through the gamma transitions of 238 keV from ^{212}Pb, 727 keV from ^{212}Bi and 911 keV from ^{228}Ac. Thorium concentration for each sample was calculated as the mean value of those gamma transitions. The ^{232}Th medium concentration for all samples of the same material was calculated as the mean value of the thorium concentration of each sample.

The ^{40}K concentration for each sample was determined from its unique 1460 keV gamma transition and the medium concentration for each material was calculated by using the same radium and thorium calculation procedures.

For the concrete beam single sample, the error was propagated from variables as peak area and efficiency.

Table 1 shows the materials measured, their average values and standard deviation for ^{226}Ra, ^{232}Th and ^{40}K concentrations. All building materials in Santo André present radioactivity concentrations within the range of values obtained by other authors [3], [4], [6], [9], [15], also listed.

^{222}Rn indoor concentrations. As always people is afraid of radiation, only seven families allowed us to install the nuclear track detectors. Even for those, some detectors were removed and we lost results. Table 2 shows the medium values and standard deviation obtained in each house. As can be observed, only for one house it was possible to assess the four seasons.

Table 1: Mean natural radionuclides concentrations (in Bq·kg^{-1}) in the building materials used in Santo André district and literature values, in brackets

Materials	Samples	^{226}Ra	^{232}Th	^{40}K
sand [a]	10	31.2 ± 1.9 (7-81)	56.6 ± 4.5 (9-104)	349 ± 10 (37-666)
cement [a]	5	53.3 ± 1.2 (1-204)	18.7 ± 0.6 (11-192)	160 ± 6 (18-555)
broken stone [a]	8	14.0 ± 0.8 (2-56)	64.3 ± 6.8 (2-93)	866 ± 52 (26-1262)
concrete [a]	10	21.5 ± 0.6 (8-146)	98.3 ± 2.2 (9-225)	1050 ± 18 (394-1856)
roofing brick [a]	2	28.8 ± 0.8 (25-160)	25.4 ± 1.2 (70-180)	332 ± 12 (550-1100)
concrete beam [b]	1	41.4 ± 2.5	58.8 ± 5.5	605 ± 36

[a] Mean concentration and standard deviation

[b] Concentration and error

Table 2: Indoor ^{222}Rn concentrations

Dwelling	^{222}Rn concentration (Bq·m^{-3})			
	Summer	Autumn	Winter	Spring
1	30.5 ± 2.1	----[a]	----[a]	----[a]
2	30.5 ± 0.7	39.5 ± 2.1	----[a]	----[a]
3	27.0 ± 1.4	33.3 ± 2.8	----[a]	----[a]
4	29.0 ± 1.4	38.5 ± 2.1	42.5 ± 3.5	40.0 ± 2.8
5	23.0 ± 1.4	27.0 ± 1.4	37.0 ± 1.4	----[a]
6	31.0 ± 1.4	36.5 ± 0.7	39.0 ± 1.4	----[a]
7	----[a]	35.5 ± 2.1	40.0 ± 2.8	36.5 ± 2.1

[a] The detectors were removed by dwellers

Indoor effective dose equivalent rate from building materials. The indoor effective dose rate by external and internal exposure due to the building materials used in the construction of the popular houses in the Santo André district of São Paulo, Brazil, was performed following the UNSCEAR [1] procedures, using the medium radionuclide concentrations of the materials (external exposure), indoor radon concentration (internal exposure) and introducing a so-called reference room concept. Ackers [16] defined a reference-room by the requirement that the effective dose rate for a person in this room is equal to the average effective dose rate experimented by a person living in a house of the type to be represented.

The effective dose rate by gamma irradiation, D_{ext}, from building materials in $mSv \cdot y^{-1}$, was calculated using the equation (1) [17]:

$$D_{ext} = p\,T\,b\,10^{-6}\,\Sigma[(q_{Ra}C_{Rai} + q_{Th}C_{Thi} + q_K C_{Ki})\,m_i] \quad (1)$$

where $\mathbf{D_{ext}}$ is the external exposure dose rate by gamma irradiation ($mSv \cdot y^{-1}$), **p** is the fraction of time spent indoors, **T** is 8760 hours per year, **b** is the conversion of absorbed dose in air to effective dose equivalent ($Sv \cdot Gy^{-1}$), **i** is the index for type of building material, $\mathbf{q_{Ra}}$ is the conversion of ^{226}Ra concentration building material to absorbed dose rate in indoor air $[(nGy/h) \cdot (Bq/kg)^{-1}]$, q_{Th} id. for ^{232}Th, q_K id. for ^{40}K, $\mathbf{C_{Rai}}$ is the ^{226}Ra activity concentration in type i of building material, $\mathbf{C_{Thi}}$ id. for ^{232}Th, $\mathbf{C_{Ki}}$ id. for ^{40}K and $\mathbf{m_i}$ is the mass fraction of type i material in reference room.

For the ^{226}Ra, ^{232}Th and ^{40}K activity concentrations we used the Table 1 experimental values. For the fraction of time spent indoors, p, and the conversion of absorbed dose in air to effective dose equivalent, b, we considered the UNSCEAR [1] values of 0.8 and 0.7 $Sv \cdot Gy^{-1}$, respectively.

For the conversion factors q_{Ra}, q_{Th} and q_K we used the Koblinger values [18] because the studied houses in Santo André had dimensions and wall thickness similar to his theoretical room model. In the calculation, Koblinger [18] considered the ^{40}K isotope and all elements in the ^{238}U (^{226}Ra) and ^{232}Th series, assuming equilibrium of the daughters and not taking into account the effect of radon emanation from the walls. The conversion factors values used were 0.79, 0.89 and 0.07 $[(nGy/h)(Bq/kg)^{-1}]$ for q_{Ra}, q_{Th} and q_K, respectively.

The effective dose equivalent rates by external exposition calculated for each building material used in Santo André are presented in Table 3.

Table 3: Indoor effective dose equivalent rate by external exposure from Santo André building materials

Building material	Indoor effective dose rate ($mSv \cdot y^{-1}$)
sand	0.17
cement	0.03
broken stone	0.14
concrete	0.31
roofing brick	0.02
concrete beam	0.004

The effective dose rate by radon inhalation, D_{int}, in $mSv \cdot y^{-1}$, was calculated using the equation (2) [17]:

$$D_{Rn} = p\,T\,r_{Rn}\,(A/v)\,(1/v)\,(0.45 - 0.15\,v)\,R \quad (2)$$

where $\mathbf{D_{Rn}}$ is the internal dose rate from radon inhalation ($mSv \cdot y^{-1}$), **p** is the fraction of time spent indoors, **T** is 8760 hours per year, $\mathbf{r_{Rn}}$ is the conversion of equivalent equilibrium concentration to effective dose per, **A/V** is surface/volume, **v** is the ventilation rate (h^{-1}), $\mathbf{(0.45 - 0.15 \cdot v)}$ is the dimensionless equilibrium factor for radon for ventilation rate in the interval of 0.1 to 2 exchanges per hour and **R** is the radon exhalation rate from dwelling.

For the fraction of time spent indoors, p, and the conversion of equivalent equilibrium concentration to effective dose per hour, r_{Rn}, we considered the UNSCEAR [1] values of 0.8 and 0.9×10^{-5} $mSv \cdot h^{-1}$ per $Bq \cdot m^{-3}$, respectively.

The relation surface/volume used was 2.0 m^{-1} according to houses dimensions. The ventilation rates were calculated according to Keller [19], through the house radon exhalation rate, the relation surface/volume and the radon concentration and ranged from 0.28 for the winter to 0.51 for the summer. The radon exhalation rate from dwellings was calculated through the radon exhalation rates from both building materials and soil underneath the construction according to UNSCEAR procedures and the value obtained was 5.91 $Bq{\cdot}m^{-2}{\cdot}h^{-1}$.

Table 4 shows the effective dose rate by internal exposure due to radon inhalation in each house at Santo André for all seasons.

Table 4: Effective dose rate from radon inhalation ($mSv{\cdot}y^{-1}$)

	Effective dose rate ($mSv{\cdot}y^{-1}$)			
Dwellings	Summer	Autumn	Winter	Spring
1	0.75	----	----	----
2	0.75	1.01	----	----
3	0.65	0.82	----	----
4	0.71	0.97	1.09	1.01
5	0.55	0.65	0.94	----
6	0.77	0.94	1.01	----
7	----	0.90	1.01	0.94

The total effective dose rate from natural gamma radiation emitted by the building materials used in Santo André was calculated by summing the results for each particular material, but the concrete beam for its relatively low value. So, the total gamma indoor effective dose received by the dwellers is 0.67 $mSv{\cdot}\ y^{-1}$, slightly lower than the value of 0.8 $mSv{\cdot}\ y^{-1}$ assumed by UNSCEAR (1988) for natural gamma radiation sources. However, we must point out the importance of knowing the contribution of naturally occurring radionuclides in building materials, since the effective dose equivalent rate due to terrestrial gammas disregarding building materials assumed by UNSCEAR (1988) is only 0.34 $mSv{\cdot}\ y^{-1}$.

The total effective dose rates for the Santo André condominium houses were assessed considering both the internal exposure due to radon inhalation and the external exposure due to gammas of building materials. The average results for summer, autumn, winter and spring were, respectively, 1.37, 1.55, 1.68 and 1.65 $mSv{\cdot}\ y^{-1}$, all below the value of 2.4 $mSv{\cdot}\ y^{-1}$ adopted by UNSCEAR (1988) for all natural radiation sources, so the houses are suitable for living.

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