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DOSIMETRY OF RADIUM EQUIVALENT IN CONSTRUCTION MATERIAL OF BRICKWORKS IN SÃO JOSÉ DO SABUGI CITY - PARAÍBA

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ABSTRACT

The earth's crust has in its composition the Naturally Occurring Radioactive Material (NORM) that may have increased concentration due to activities of exploration and extraction of environmental resources. The civil construction is an economic activity that requires the use of much of the natural resources, such as the raw material of brickworks, like clays, mainly used for the production of bricks and tiles. These construction materials may contain high levels of natural radioactive elements, even with concentrations higher than the limits established, given that the levels vary according to the composition of rocks and soil, due to the geological formation and may result in increased exposure of humans to natural radioactive activities. In this context, the radioecologic dosimetry is defined in terms of Radium Equivalent activity (Ra_{eq}), that ensure radiometric conditions for the use of material derived from clays before its final application in housing construction, an initiative that ensures the radioecologic safety of population. Thus, this study aimed to establish the calculation of Ra_{eq} in the raw material of brickworks located in São José do Sabugí city, state of Paraiba, in an area adjacent to the uranium deposits of Espinharas, to estimate the risks associated with primordial radionuclides attributed to TENORM activities (Technologically Enhanced Naturally Occurring Radioactive Materials) from the extraction and use of clay as a raw material in the manufacture of bricks and tiles. Analyses were performed by High Resolution Gamma Spectrometry, with HPGe-Be detector, assuming the state of secular radioactive equilibrium. The results ranged from 183.2 to 747.78 Bq/kg, with an average of 494.6 Bq/kg which exceeded the limit of 370 Bq/kg established by UNSCEAR for construction materials. Some samples obtained values exceeded by up the double this limit, suggesting control and radiometric certification for application of this material.

Keywords: natural radioactivity; gamma spectrometry; equivalent radio, construction.

1. INTRODUCTION

Chemical elements may present spontaneous mass loss property from the energy emission and are characterized as radionuclides. The emitted radioactivity is defined as natural when occurs spontaneously and can be emitted as particulate form, such as alpha and beta particles and through electromagnetic radiation, as gamma rays [7].

The knowledge of the distribution and dynamics of various types of natural radionuclides in terrestrial environments is an important aspect of the radioecology study. In different types of soils and rocks, environmental radioactivity is coordinated by emission of energy for a variety of chemical elements, which creates a continuous natural radiation exposure of the general population. In this context the behavior and transfer routes of ^{226}Ra and their decay product, ^{222}Rn, attracts interest particular, because this correspond about 50% of the human radiation exposure dose [6, 3].

Of the all radioisotopes, stand out the decay products of the ^{232}Th and ^{238}U series, being from 100% of natural thorium and 99.27% of the isotopic composition of natural uranium, respectively. These minerals are found in the earth's crust, which can be extracted for use in various activities, in this case, unhealthy, because carry a variety of radioactive energy sources [3].

The terrestrial radioisotopes have not homogeneous distribution along the crust, just because exist places with higher concentrations that other places. The existence of high levels of radioactive minerals in some areas of the earth's crust enables its transition to areas of high human habitation during the exploration process [1].

The man, by survival principles, began the use of natural resources for your comfort. Thus, for thousands of years, develop ceramic manufacturing activities, represented by pots, kitchen utensils and, later, materials for construction, such as bricks and tiles. These types of materials can promote considerable radioactivity exposure, depending on the source of raw material used in manufacturing [14].

Clays are the main raw material used in the ceramics production, especially the red clay for the volume of production and demand [9]. Brazilian red clay reserves are large and distributed in all regions of country [2].

In Brazil, the regulation imposed and active legislation are consistent for direct exposures to workers, such as mineral exploration, diagnostic activities in radiology clinics or nuclear power plants, not promoting enough attention to possible exposures to radon and other radioactive sources in inside civil constructions [4].

In this context, this study aimed to estimate the calculation of radium equivalent activity (Ra_{Eq}) to be used for measure the ambient dose associated with primordial radionuclides attributed to Technologically Enhanced Naturally Occurring Radioactive Materials activities (TENORM) from the extraction and the use of clay as a raw material in the manufacture of bricks and tiles in brickworks located in the São José do Sabugí city, adjacent to the uranium mine of Paraíba of São José de Espinharas city.

2. MATERIAL AND METHOD

Eight clay samples were collected in five brickworks distributed in the area of São José do Sabugí city, State of Paraíba. Were collected approximately 3.0 kg of each sample of the raw material used to produce bricks and tiles that were packed in thick plastic bags, sorted by company (brickwork) and local of extraction.

The samples were prepared for spectrometric analyzes, by the following procedures: (a) drying in stove at 60 °C; (B) fragmentation; (C) reduction of particle size to passing the mesh of 1.0 mm ; (D) homogenizing the sample and weighed (250g per sample); (E) packaging in polyethylene containers hermetically sealed and marked. It's worth noting that some of the material that was retained on the sieve during the sieving process was macerated with wooden roller so that it could reach sufficient particle size for its passage in the 1mm sieve diameter.

After treatment and considering that the method of analysis requires the establishment of secular radioactive equilibrium in the evaluation of ^{226}Ra and ^{232}Th, it was expected to at least 40 days with the sealed samples for analysis begins in gamma spectrometry system.

It was used the hyperpure germanium detector (HPGe) of high resolution gamma spectrometry by Canberra® mark. It was needed ^{241}Am, ^{133}Ba and ^{152}Eu sample standards, with initial activities certified by the Instituto de Radioproteção e Dosimetria (IRD) - Rio de Janeiro, Brazil, to determine the ^{226}Ra, ^{232}Th and ^{40}K, and construction of the counting efficiency curve. Following the basis of suggestions from IAEA (1991), from TECDOC-619 technical document, for calibration of the measurement system, major transitions range of sealed sources of ^{241}Am, ^{137}Cs and ^{60}Co were used.

During the calibration process, the sources were individually measured in a time of 3,600 seconds. After obtaining the data, there was a qualitative adjustment depending on the analyzed energy sources and channels available in the electronic system. The equation 1 enabled identification of photopeaks, where "x" represents the correlation between the center channel and the MCA and "E" represents the energy of the gamma photons interacting within the detector. The obtained calibration straight line can be observed in the Figure 1.

$$E = 0{,}4456(x) - 1{,}553 \quad (1)$$

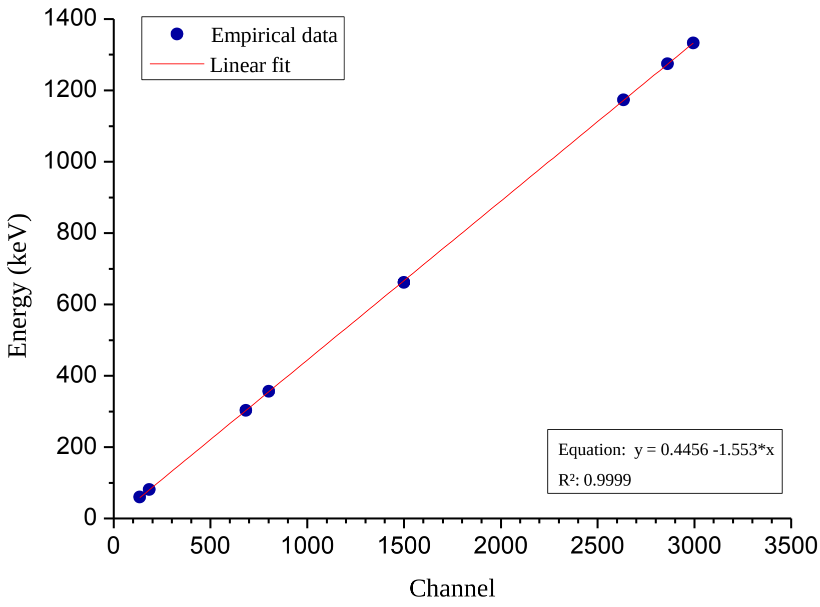

Figure 1: Measurement System straight calibration.

For the efficiency curve construction (Figure 2), the standard sources constituted arrays similar to brickworks samples where realized the doping with low radiation considered previously analyzed sources. Then, the arrays were dried at 40 °C, homogenized, packed in polyethylene containers and measured individually, with standardized acquisition times of 3,600 seconds. The efficiencies for each energy and the results were plotted on a statistical program for the preparation of a semi-empirical model explaining the efficiencies behavior.

The energies were selected by the 619 technical document (TECDOC 619) of the International Atomic Energy Agency [5]. The corrections of the activities of standards were calculated by Equation 2, where "A_{Eip}" is the activity to be corrected, based on the initial activity "A_{Eip0}" and radioactive decay constant "λ".

$$A_{Eip} = A_{Eip_0} \times e^{-\lambda \times t} \quad (2)$$

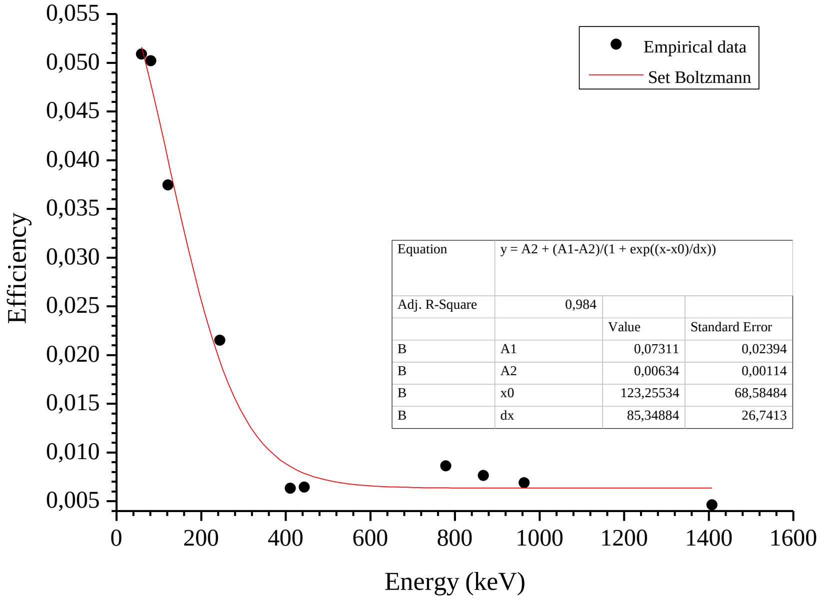

Figure 2: Measurement System efficiency curve.

To perform a statistical study of the results obtained for the average activity of the radium equivalent, was used the BioEstat 5.0 program to determine the descriptive statistics of the data. To determine the normal tendency in the distribution of data, nonparametric test of Shapiro-Wilk was applied, with 95% significance level.

According to Equation 3 below, where C_{Ra}, C_{Th} and C_K correspond to specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, expressed in $Bq.kg^{-1}$, it was obtained to calculate the Equivalent Radium Activity to estimates the resulting activity of gamma emitters of the main contribution of these series.

$$Ra_{Eq} = C_{Ra} + (1.43 \times C_{Th}) + (0.077 \times C_K) \quad (3)$$

3. RESULTS

The obtained results revealed the radium equivalent activity values ranging from 183.2 to 747.78 Bq.kg^{-1}, featuring considerable dispersion, with the measure of central tendency value of 494.6 expressing the radiometric situation area; the standard deviation obtained was 205.8. The diagram showing in the Figure 3 allows to note, considering the standard deviation associated with each value, that the maximum value may reach 791.2 Bq.kg^{-1}, reinforcing the relevance of the proposed dispersion when considering already that the lower value may reach 161.6 Bq.kg^{-1}.

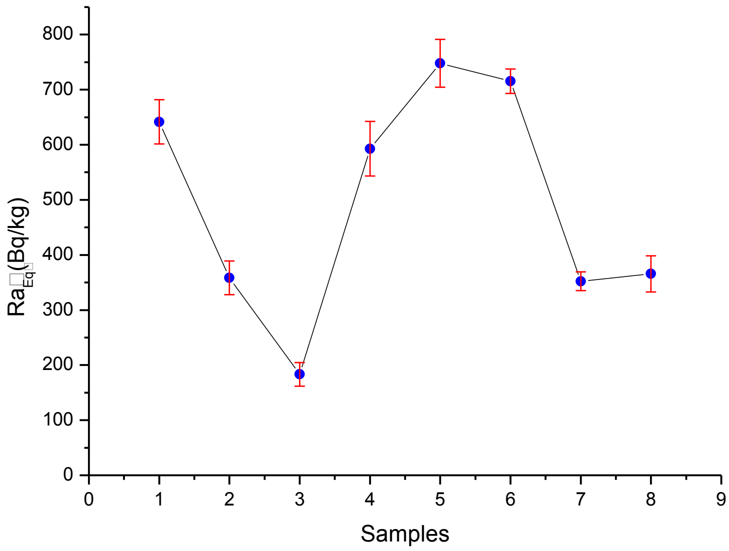

Figure 3: Radium equivalent activity and uncertainties diagram.

Considering the average value of radium equivalent defined by the IAEA and UNSCEAR (370 Bq.kg^{-1}) for building materials, it can be said, in this research, that the average obtained from the statistical analysis, with a 95% confidence level, referencing the municipality in this study consisted be higher, still being considered almost 5 times higher than the world average (104.2 Bq.kg^{-1}).

The diagram showed in Figure 4, builded by the presence of outliers data, enables accurate representation of the minimum and maximum values, 183.2 and 747.78 Bq.kg^{-1}, respectively, still exhibiting a range with approximately 564.58 Bq.kg^{-1}. The point corresponding to the maximum value also shows the presence of some outliers. The graphical difference between the average and median values show the distribution abnormality and the variation in the relation between the median and quartiles shows the influence of some values on the results.

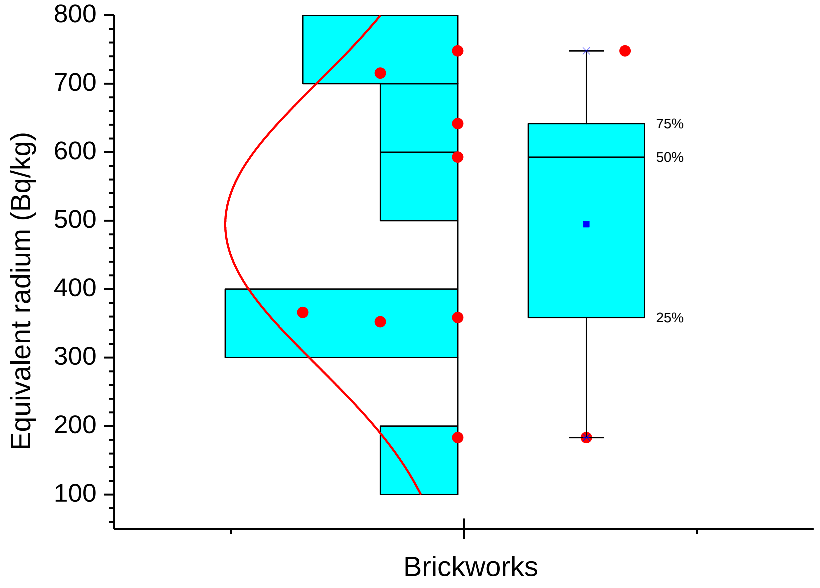

Figure 4: Distribuition values of the Ra_{Eq}.

The Table 1 shows the results obtained in this study and other studies conducted in Brazil and in some countries, and the world average associated with radium equivalent in soil.

Table 1: Comparative study of radium equivalent activity.

STUDY AREA	Ra_{Eq} ($Bq.kg^{-1}$)	REF
São José do Sabugí	494,6 (183,2 - 747,8)	This Study
Soil/Pedra-PE	319,2	[12]
RG/Pedra-PE	327,5	[12]
RCS/Pedra-PE	70.124,5	[12]
Rock/São José de Espinharas-PB	3.672,6	[16]
World average	104,2	[10;13; 11; 19]
Taiwan	96,1	[17]
Coastal Kartanaka	53,9	[13]
Tripoli	34,4	[15]
Dhaka City	67,1	[13]
Ireland	124,1	[8]
Malaysia	75,4	[19]

Ra_{Eq}: Radium equivalent activity; RCS: Calcium-silicate rocks; RG: Granitic rocks.

In studies conducted in Brazil, precisely in the state of Pernambuco, Santos Junior (2009) found, from studies in rocks and soil of the Pedra city, Ra_{Eq} values of 327.5 $Bq.kg^{-1}$ to granitic rocks and 70,124.5 $Bq.kg^{-1}$ for amphibolitic calcium-silicate rocks analyzed that still showed higher contribution of the specific activity of ^{226}Ra to calculate Ra_{Eq}, corroborating with this research. In the same study, to the ground analyses, obtained 319.2 $Bq.kg^{-1}$ average for the radium equivalent activity, lower than the average of this study and also lower than the limit established by UNSCEAR. All these results are shown higher than world averages.

With more effective contribution to the results of this study, in consideration of analyzed area, research conducted by Silva (2014), which examined rocks from the city of São José de Espinharas-PB , near to the Paraíba uranium deposit and also near to the area studied in this research, obtained average value of 3,672.6 $Bq.kg^{-1}$ for the radium equivalent activity, considered with great significance, and about 10 times higher than the proposed limit by UNSCEAR and more than 7 times higher than the average obtained in this study.

The estimated world mean values of radium equivalent activity promotes the understanding regarding the distribution of radionuclides in the earth's crust at varying levels [18]. Among the results of world research cited, there are similarities with respect to this research when considering the lower contribution of specific activities of ^{40}K in the calculations of radium equivalent activity, except in research realized by Shenber (1997) in Tripoli and Selvasekarapandian *et al.* (2000) in Dhaka City, where the ^{232}Th, with activities from 9.5 and 16.0 $Bq.kg^{-1}$, respectively, set less contribution to the calculations of Ra_{Eq}.

It is observed that the calculations of specific activities as well as the radium equivalent less discrepant in relation to the limits established, are in Tripoli, established in searches made by Shenber (1997), where, with radium equivalent activity of 34.4 $Bq.kg^{-1}$, is characterized as the lowest value to be compared, more than 14 times lower than the average found in this research, followed by Coastal Kartanaka and Dhaka City [13] and Malaysia [19] at 53.9, 67.1 and 75.4 $Bq.kg^{-1}$, respectively, as the lowest results, characterizing the areas with normality radioactive standards.

4. CONCLUSIONS

The results of this study showed that the analyzed area presents some values for equivalent radium activity above the limits established by the certification entities.

The spectrometric analysis showed that 50% of the analyzed area presents levels of natural radioactive elements above the limit established by UNSCEAR reaching values up to 2 times higher than those considered normal.

From reported data, the São José do Sabugí city requires specific intervention, which a monitoring action of the supply sources of raw material for the brickworks, as well as the final products (tiles and bricks) because were evidenced in some of its samples, very high values for the analyzed radionuclides.

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