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Activity concentration of ^{226}Ra, ^{232}Th, ^{40}K and ^{222}Rn in soil samples of the Campos Gerais region (Paraná)

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ABSTRACT

The aim of the present work was to determine activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{222}Rn concentration in soil samples from Telêmaco Borba, Tibagi, Curiúva, Ventania, Lageado Bonito and Ortigueira cities. The soil samples, with about 2 kg, were collected on the surface at points near the river bed, making sure that the soil is not under the agriculture influence. Gamma-ray spectrometry analyses were performed with a NaI(Tl) scintillation detector, with dimensions of 76 mm in diameter, 152 mm in height and 8 cm of lead shield. The ^{222}Rn concentration determinations were performed by means of the sealed can technique, using as track detector a polycarbonate of domestic manufacture. The results obtained for the activity concentration varied from <4.2 ± 1.2 Bq.kg^{-1} to 290.2 ± 1.7 Bq.kg^{-1} for the ^{40}K, from 23.3 ± 1.1 Bq. kg^{-1} to 36.5 ± 1.1 Bq.kg^{-1} for the ^{226}Ra and from 52.6 ± 1.9 Bq.kg^{-1} to 70.5 ± 2 Bq.kg^{-1} for the ^{232}Th. The results for the ^{222}Rn concentration ranged from 4 ± 3 Bq.m^{-3} to 1147 ± 326 Bq.m^{-3}. Two of the six soil samples presented radon concentrations above the threshold established by the World Health Organization of 200 Bq.m^{-3}. The concentrations of ^{226}Ra and ^{232}Th are within the limits, showing the importance of future investigation on the origin of this radon.

Keywords: **Soil, Radon, Natural radioactivity, Campos Gerais Region**

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1. INTRODUCTION

1.1. Natural radioactivity

It is known that the human species is constantly exposed to radiation sources of natural and artificial origin and that sources of natural origin, generally, represent the greatest contribution from the dose received. Exposure of natural origin comes from space (cosmic radiation) and from radionuclides present in soil, building materials, water and air or food [1].

The intensity of this exposure varies according to geographic location and depends on many factors such as soil composition, climatic conditions and socio-cultural habits [2].

Many studies have been carried out around the world to assess the concentrations of natural radionuclide activity in soils. These types of works aim to, besides generating a database regard the regions not yet studied, evaluate risks to the population [3], verify the local geological influence on the level of radioactivity [4-6], and assess the influence of agriculture on radiation levels [7]. In Brazil, several works were carried out with the same objectives mentioned [8-12].

In Paraná, there are some studies evaluating radioactivity in soils in the city of Figueira, as it has an active coal mine [13-14], and some punctual works evaluating soils in the city of Londrina [15]. In the area studied (located in the region called Campos Gerais), there is no data on natural radioactivity in soils. This type of survey is important because in the geological context of the region, sedimentary basin of Parana, there are some sedimentary formations favorable to the occurrence of U and Th [16-17].

The objective of this study was to determine the activity concentrations in soil samples from six cities (Tibagi, Lageado Bonito, Ortigueira, Telêmaco Borba, Ventania and Curiúva) located in Campos Gerais region- Paraná, Brazil.

1.2. Radon

The presence of radon and its daughters in homes is seen by most researchers as a risk factor for public health [18]. In several countries there are researches with the objective of determining the average concentration of radon (mainly the isotope 222) and daughters which provides subsidies for conducting epidemiological studies that correlate concentrations of ^{222}Rn and daughters with the incidence of deleterious cancer effects mainly on the respiratory system [2].

^{222}Rn, an isotope of radon, that originates from the radioactive decomposition of ^{226}Ra by alpha decay in the ^{238}U radioactive series, being the only gaseous element in this long series of radioactive decays. Some of its main features are the difficulty of interacting with other elements, the ability to emanate from the soil or rock with extreme ease and the ability to concentrate indoors [19].

As uranium and radio are always present in soil, rocks and water, it is natural the presence of radon in the air, which, in open air environments presents low concentration due to its continuous dispersion in the atmosphere. However, in closed and poorly ventilated environments the uranium and radio concentrations can reach high values.

According to Alberigi (2006), the interest in determining the radon concentrations is due to the fact that its inhalation is associated with the incidence of respiratory diseases [1]. The earliest reports date to the 16th century, when mining regions in Germany became known by the incidence of a disease that was known as mountain sickness, now known as lung cancer [20].

The increase in uranium mining in the 1940s caused an increase in lung cancer cases in miners, which consequently led to the association between exposure to radon and lung cancer [20]. Currently, according to a report published by the World Health Organization, it is known that radon is the second largest cause of lung cancer in the world, second only to tobacco [19].

Radon gas decays to several daughters, which are also radioactive. For the purpose of contributing to the radiation exposure, the major importance is concentrated in the first four daughters of short half-lives, ^{218}Po (3.05 min), ^{214}Pb (26.8 min), ^{214}Bi (19.9 min) and ^{214}Po (0.000164 s). As ^{218}Po and ^{214}Po are alpha emitters, whose linear transfer of energy to alpha particles is 20 times greater than for reference radiations (electrons), are more detrimental and responsible for the increased incidence of lung cancer [21].

1.3. Gamma radiation

Gamma rays are a form of electromagnetic radiation with a wavelength between 10^{-14} and 10^{-8} m. These rays are emitted during radioactive decay of elements usually present in all soils and rocks, regardless of small quantities as trace elements.

The main sources of natural radiation are the ^{40}K, ^{238}U and ^{232}Th series. In the Earth's crust, ^{40}K has activity concentrations ranged from 623 Bq.kg^{-1} to 783 Bq.kg^{-1}, ^{238}U from 24.7 Bq.kg^{-1} to 37.05

Bq.kg^{-1}, and ^{232}Th from 32.48 to 48.72 Bq.kg^{-1} [22]. The ^{40}K corresponds to 0.012% of the total potassium, ^{238}U corresponds to 99.3% of the total uranium, and ^{232}Th to 100% of the total thorium [22].

Gamma spectrometry measures the activity concentration of ^{40}K, ^{238}U and ^{232}Th in rocks and soils by detecting and quantifying the natural gamma radiation emitted by them or their daughters. The ^{40}K emits gamma rays when it decays to ^{40}Ar and the radiation energy emitted by ^{40}K is 1.46 MeV [23].

The ^{238}U and ^{232}Th concentrations are more complex measures because ^{238}U and ^{232}Th decay through a series of daughter nuclides until they reach stable Pb isotopes. In this work, we determined the activity concentration of ^{226}Ra instead of its parent ^{238}U. The energy of the diagnostic radiation of ^{226}Ra is 1.76 MeV, which is associated with the ^{214}Bi nuclide. The energy of the diagnostic radiation of ^{232}Th is 2.62 MeV, which is associated with the ^{208}Tl nuclide [23].

Gamma spectrometers are equipment that separate energy from gamma radiation into windows or banners. The detector, usually a thallium-activated sodium iodide crystal (NaI(Tl)), absorbs gamma radiation and turns it into light pulses or scintillations. These scintillations are converted into electrical signals proportional to the intensities of light. Then, the signals are separated into classes, depending on the magnitude, in order to obtain an energy spectrum of the incident gamma rays. A calibration process allows the amount of radiation with energies of 1.46 MeV, 1.76 MeV and 2.62 MeV, measured in scintillations per second (cps), to be expressed as activity concentration of ^{40}K, ^{226}Ra and ^{232}Th respectively.

2. MATERIALS AND METHODS

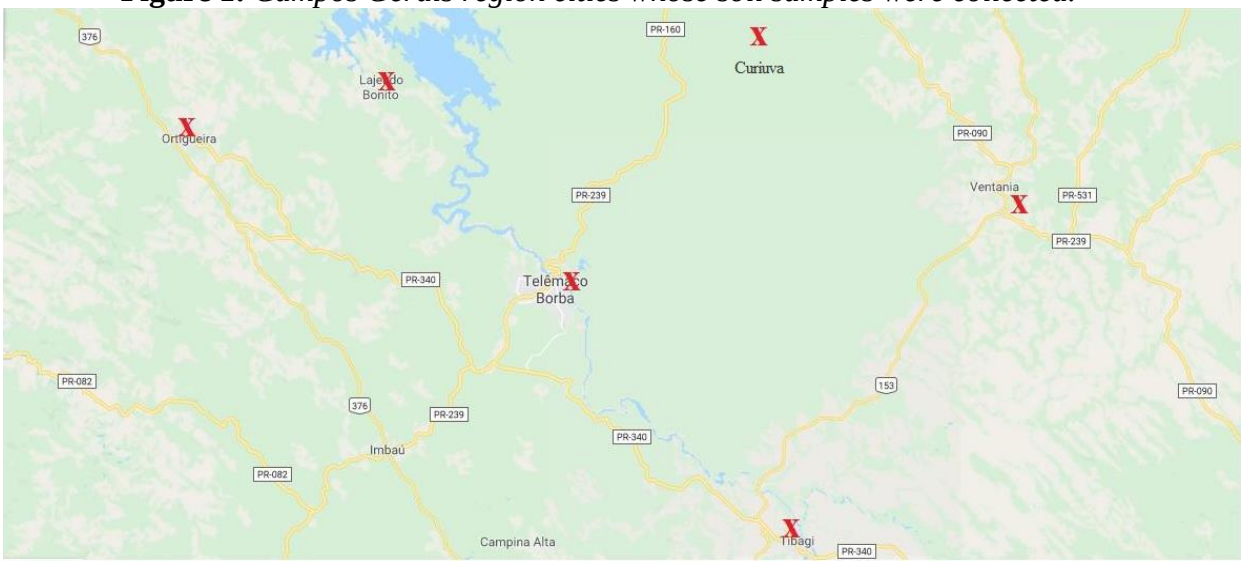
2.1. Study area and sample collection

The soil samples were collected in six cities of the Campos Gerais region, near the city of Telêmaco Borba. These samples were coded as shown in Table 1. The Figure 1 shows a map with the cities where the samples were collected, at the river banks in preserved areas.

Table 1: Codes of the soil samples and sampling sites

Sample Code	City
1	Tibagi
2	Lageado Bonito
3	Ortigueira
4	Telêmaco Borba
5	Ventania
6	Curiúva

Figure 1: *Campos Gerais region cities whose soil samples were collected.*



Source: Adapted from Google Maps

The soil samples, which had a mass of approximately 2 kg, were obtained at topsoil at depths of approximately 20 cm, in August 2018, and stored in plastic bags. In the laboratory, the material was spread on a tray to dry. After drying, the samples were pulverized and homogenized, and divided into aliquots that were weighed and hermetically sealed in the polyethylene containers. The gamma and radon measurements were performed after 30 days (to allow the equilibrium of ^{226}Ra and ^{232}Th with their respective short half-life daughters). This condition is necessary because ^{226}Ra and ^{232}Th are determined from daughters of ^{222}Rn and ^{220}Rn respectively and these are noble gases easily

escape from the samples, needing to be sealed long enough for the radioactive equilibrium in both series to be reestablished [24].

2.2. Determination of radon concentrations

The determination of the radon activity concentration was performed using solid nuclear track detectors (SSNTD) by means of the Sealed can technique using a polycarbonate of domestic manufacture, as solid detector of nuclear tracks [25].

This technique consists in sealing a certain mass of the samples in a cylindrical polyethylene vessel with a diameter of 0.095 m and height of 0.15 m. In this work, each sample weights 80 g. This sample should cover the entire bottom of the vessel and the SSNTD detector should be attached to the top of the vessel. The measurements were carried out in triplicate, and to assure accurate background measurements, empty containers were used. The radon concentration is then determined by equation 1 [26, 27]:

$$C_{Rn} = \frac{D}{K.t} \quad (1)$$

Where, C_{Rn} is the concentration of ^{222}Rn in $Bq.m^{-3}$; D is the density of tracks in $tracks.cm^{-2}$; t is the exposure time in *days*, the time the vessel has been sealed, and K is the trace detector efficiency given in $tracks.cm^{-2}/Bq.m^{-3}.days$. In this work, the density was obtained by using manual counting of tracks with optical microscopy. The Physis binocular microscope had an integrated camera with a viewing resolution of 1024 X 768 pixels and an image capture resolution of 2048 X 1536 pixels.

In order to determine the radon concentrations in the air, it is necessary to know the detector's trace density, exposure time, and the K-efficiency of the detector used. This efficiency, also called the "radon concentration trait conversion factor" or simply "calibration factor", allows the conversion of track density into radon activity.

The K-efficiency is determined from equation 1 when the ^{222}Rn concentration supplied by the calibrated source, the detector exposure time and the track density in the detectors are known. The calibration factor is expressed in $tracks.cm^{-2}/Bq.m^{-3}.days$. In this work, it was adopted the value of K 1.60 ± 0.10 $tracks.cm^{-2}/Bq.m^{-3}.days$, which was obtained by Aquino (2015) for the same detector

in similar experimental conditions [28]. As this is a methodology implementation phase, the participation in proficiency tests has not yet been carried out.

2.3. Determination of gamma activities

For these analyzes the samples, with a mass of 140 g were sealed for 30 days in a cylindrical polyethylene containers and were measured for a period of 24 hours.

The analyses were performed using a gamma spectrometer from Amptek (GammaRad5 model), with the following specifications: the dimensions of crystal were 76 mm in diameter and 152 mm in height and NaI (Tl) scintillator detector coupled to a photomultiplier and digital pulse processor (DP5 - Amptek). The measuring system is protected by an 8 cm lead shield.

The peaks used for radioisotope concentration estimates were 2614 keV of ^{208}T1 (^{232}Th series), 1120 keV of ^{214}Bi (^{238}U series) and 1461 keV of ^{40}K. Each sample was measured for 24 hours.

The efficiency calibration was accomplished by the method indicated in the IAEA's TECDOC-1363 [23]. In this procedure, the radiation from the unknown sample is compared with the radiation from known standards. The method considers the background-corrected count-rates of the spectrum's regions of interest (ROIs) and strips the cross-influence among ROIs. As reference materials, it was used the IAEA reference standards for the analysis of radioisotopes in geological materials, known as RG-Set. The RG-Set is a set of three samples, RG-K-1, RG-Th-,1 and RG-U-1, with known and relatively high concentrations of potassium, thorium and uranium, respectively. RG-U-1 and RG-Th-1 are uranium and thorium ores diluted in the high purity silica matrix and RG-K-1 is potassium sulfate with a high degree of purity. According to the IAEA Certification Report, the ^{238}U and ^{232}Th series are in equilibrium in the RG-U-1 and RG-Th-1 samples.

3. RESULTS AND DISCUSSION

3.1. Radon activity concentrations

The results of the average of the radon determination triplicates of each sample, with the respective propagated uncertainty, are shown in Table 2. The track counts were performed manually

after exposure to the natural radiation detector of the samples and treatment for the revelation of tracks [25].

Table 2: Radon activity concentrations in soil samples from Campos Gerais region

Sample Code	C_{Rn} (Bq.m^{-3})
1	8 ± 4
2	4 ± 3
3	46 ± 15
4	562 ± 179
5	192 ± 43
6	1147 ± 326

In Table 2 it can be observed that the uncertainties associated with samples 3, 4, 5 and 6 were around 30%, an acceptable result considering the technique used and the fact that this technique is in the implementation phase. Furthermore, samples 4 and 6 showed a concentration above the limit (200 Bq.m^{-3}) established as safe by WHO (World Health Organization) [19]. This is an indication that more study should be done at these two sampling points.

3.2. Concentrations of gamma activity

In the activity concentration measurements through gamma ray spectrometry with NaI(Tl), the minimum detectable concentrations (LLD), calculated as in [29], were 4.2 Bq.kg^{-1} for ^{40}K, 3.4 Bq.kg^{-1} for ^{226}Ra and 5.7 Bq.kg^{-1} for ^{232}Th. The results obtained for the activity concentrations are shown in Table 3.

Table 3: Activity concentration in soil samples from Campos Gerais region. The uncertainties presented are the propagated count-rates standard deviations.

Sample Code	^{40}K (Bq.kg^{-1})	^{226}Ra (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})
1	<4,2	23 ± 1	53 ± 2
2	264 ± 2	36 ± 1	69 ± 2
3	111 ± 1	34 ± 1	71 ± 2
4	290 ± 2	29 ± 1	54 ± 2
5	126 ± 1	24 ± 1	67 ± 2
6	76 ± 1	25 ± 1	65 ± 2

The values presented in Table 3 are within the range of values normally found in the soil, as described in the worldwide literature, with an average value of 35 Bq.kg^{-1} for the ^{232}Th, 30 Bq.kg^{-1} for the ^{226}Ra and 400 Bq.kg^{-1} for the ^{40}K [21]. In the national literature, similar values were also found by Leal et al. (2020), in soils in the state of Pernambuco, with an average value of 37 Bq.kg^{-1} for the ^{232}Th, 62 Bq.kg^{-1} for the ^{226}Ra and 464 Bq.kg^{-1} for the ^{40}K [12]. Ribeiro et al. (2018) evaluated soils in the state of Rio de Janeiro and obtained average values of 74 Bq.kg^{-1} for the ^{232}Th, 32 Bq.kg^{-1} for the ^{226}Ra, and 114 Bq.kg^{-1} for the ^{40}K [9]. In the state of Paraná Rodrigo et al. (2005) measured soils in the city of Londrina and obtained average values of 21 Bq.kg^{-1} for the ^{232}Th, 8 Bq.kg^{-1} for the ^{226}Ra, and 73 Bq.kg^{-1} for the ^{40}K [15].

4. CONCLUSION

The activity concentrations of the soils measured by gamma spectrometry technique were within the limits normally presented in the literature for soil samples.

Under the conditions of the present study, radon concentrations showed that for two cities (Telêmaco Borba and Curiúva) the values are above the limit established by the World Health Organization.

There was no correlation between ^{226}Ra concentrations and radon concentration in the samples studied. This is probably because the correlation between the ^{226}Ra concentration present in a geological sample and its radon emanation rate depends on some factors such as sample

mineralogy, density and porosity. Further measurements should be made of the physical and mineralogical properties of these samples for the continuation of the present study.

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