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Application of a semi-empirical physical-mathematical model in determining the activity concentration of ^{226}Ra in granites

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ABSTRACT

This study aimed to estimate the activity concentration of ^{226}Ra in granite samples collected from various regions of Brazil, using a simplified semi-empirical physical-mathematical model. The importance of such an assessment lies in the fact that granite, widely used in construction, can be a source of radioactive contamination due to the presence of ^{226}Ra, a precursor of ^{222}Rn. ^{222}Rn and its short-lived decay products are responsible for approximately 55 % of the internal radiation dose received by humans, and the emission of alpha particles by these radionuclides increases the risk of bone and lung cancer. To this end, indirect measurements of the activity concentration of ^{222}Rn were performed using the AlphaGUARD detector, a rapid, convenient, and relatively inexpensive approach compared to traditional methods. The ^{226}Ra activity concentrations obtained for the analyzed granite samples were 44.4 ± 7.2 Bq/kg, 23.4 ± 8.0 Bq/kg, 24.6 ± 7.8 Bq/kg, and 15.0 ± 5.1 Bq/kg. These results are below the reference levels established by national and international radiological protection agencies, indicating that the analyzed granites can be used without restrictions. The study was conducted in a laboratory, with samples provided by a marble company in Palmas, Paraná, Brazil. The conclusions reinforce the safety of using these materials, but also highlight the importance of continuous monitoring.

1. Introduction

Human exposure to ionizing radiation is a daily reality, originating from various natural sources. Background radiation consists of radionuclides belonging to the natural decay series of uranium (^{238}U) and thorium (^{232}Th), present in the Earth's crust, rocks, soil, groundwater, and building materials, in addition to cosmic radiation. Potassium (^{40}K) is another naturally occurring radionuclide that contributes to this exposure (UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, 2008; Eisenbud and Gessel, 1997; NNDC – National Nuclear Data and Center; Spacov, 2016; Hajj, 2017). Approximately 80 % of the ionizing radiation humans receive throughout their lifetime comes from natural background radiation, while the remaining 20 % corresponds to the sum of all artificial sources of exposure (UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, 2008; Eisenbud and Gessel, 1997; NNDC – National Nuclear Data and Center; Spacov, 2016; Hajj, 2017).

The global population is exposed to ionizing radiation in two ways: (1) external exposure, primarily related to the interaction of gamma and cosmic radiation with the external part of the human body; and (2) internal exposure, which involves the interaction of radiation with the interior of the body, caused primarily by the inhalation of radon gas. This, along with its short-lived decay products, interacts harmfully with the cells of the human respiratory system (UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, 2008; WHO – World Health Organization, 2004).

According to UNSCEAR (2008), humans receive an average annual natural radiation dose of 2.4 mSv. Of this total, 1.26 mSv results from the inhalation of radon gas and its fast decay products; 0.48 mSv is attributed to external gamma radiation; 0.39 mSv to cosmic radiation; and 0.29 mSv to the ingestion of contaminated food and water.

There are more than 30 known isotopes of radon gas, and it is the primary source of exposure to natural ionizing radiation, accounting for approximately 55 % of the total from this source. Among the isotopes, ^{222}Rn is the most radiologically relevant because, when inhaled, it is difficult to expel, being approximately nine times denser than

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atmospheric air. The concentration of this radioactive gas is directly related to the concentrations of ^{238}U and ^{226}Ra in the soil, which, in turn, depend on several factors, such as the type of soil, the nature of the rock (igneous, metamorphic, or sedimentary) that composes it, and the size of the soil grains (UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, 2008; WHO – World Health Organization, 2004; Cinelli et al., 2017).

^{222}Rn has specific characteristics: it is odorless, tasteless, and colorless, which allows its presence in indoor environments to go unnoticed. An effective way to mitigate its concentration in these environments is to keep the area ventilated. This gas is formed during the radioactive decay of ^{226}Ra and has a half-life of approximately 3.8 days. Upon decay, it emits an alpha particle (α) with an energy of 5.49 MeV. Alpha radiation has a high Linear Energy Transfer (LET) (UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, 2008; Spacov, 2016; WHO – World Health Organization, 2004).

^{222}Rn, along with its progeny, is highly toxic from a radiological point of view, as it decays by alpha emission. LET is defined as the average energy deposited per unit path length of a charged particle in a material. Thus, when interacting with lung cells, these particles transfer a large amount of energy, significantly increasing the likelihood of developing lung cancer (UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, 2008; Spacov, 2016). The precursor of ^{222}Rn, ^{226}Ra, is an alkaline earth metal with a half-life of 1600 years that, upon decay, emits alpha particles with an energy of 4.8 MeV. Due to its similar chemical properties to calcium (Ca), both belonging to group II of the Periodic Table, ^{226}Ra can be incorporated by the human body in cases of calcium deficiency, increasing the risk of developing osteosarcoma, a type of bone cancer (UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, 2008; WHO – World Health Organization, 2004).

Construction materials are a significant source of exposure to ^{226}Ra and ^{222}Rn, as they come from the processing of soils and rocks from the Earth's crust that contain significant concentrations of ^{238}U, a direct precursor of both radionuclides (Campos and Pecequilo, 2003; Corrêa et al., 1993; Narloch et al., 2019). Granite, an igneous rock widely used in construction, is used for both exterior facades and interior elements such as countertops, flooring, and wall coverings in residential and commercial settings. For its commercialization, raw granite is extracted from quarries, processed in beneficiation plants, and subsequently distributed to the consumer market (Hajj, 2017; Neves, 2005).

During this process, the radioactive equilibrium between radionuclides belonging to the same family is often disrupted, resulting in an increase in the specific activity of certain radionuclides within the radioactive series (Campos and Pecequilo, 2003; Corrêa et al., 1993; Narloch et al., 2019).

The International Commission on Radiological Protection (ICRP, 1993) establishes reference limits for the concentration of ^{222}Rn activity in indoor environments: up to 200 Bq/m^3 is considered a safe level; between 200 and 400 Bq/m^3, caution is recommended; between 400 and 600 Bq/m^3, intervention is advised; and concentrations above 600 Bq/m^3 require immediate corrective action.

During the extraction, processing, and marketing of granite, workers and consumers may be exposed to radiation emitted by the radionuclides ^{226}Ra and ^{222}Rn. Therefore, it is essential to monitor their activity concentrations, ensuring they are within the limits defined by national and international regulatory agencies.

Recent work has explored the measurement of radioactive gases and natural radioactivity in different materials and environments. Gümbür (2024) investigated the measurement of radium and radon in bottled mineral waters. Küçükönder et al (2023a) and Küçükönder, et al (2023b) addressed natural radioactivity in soil samples and the measurement of radon gas in soils from specific regions of Turkey, respectively. These studies reinforce the importance of monitoring radionuclides in various materials and environments. This study aims to infer the activity concentration of ^{226}Ra in granite samples based on the activity concentration of ^{222}Rn, to verify whether the levels of these radionuclides are within the safe limits established by national and international regulatory agencies. The originality of this work lies in the application of a simplified semi-empirical physical-mathematical model for the indirect determination of ^{226}Ra concentration, offering a faster and more economical alternative to traditional methods. The choice of granites from various regions of Brazil is justified by their importance in the production and marketing of granite worldwide, making them a strategic location for assessing potential exposure to these materials. This study makes a significant contribution to assessing the radiological risk associated with the use of granite in civil construction, providing important data for protecting public health.

2. Materials and methods

2.1. Sample preparation

The different types of granite analyzed in this study were supplied by Marmoraria Pato Bicho da Pedra, located in the city of Palmas, state of Paraná, Brazil. Samples were collected in May 2022, following the standards established by NBR 15845–1:2010 to ensure representativeness and quality. Sample selection was based on size and mass, with no additional criteria adopted. The samples, initially irregular, were processed by the supplier and made available in the form of slabs measuring 12 cm × 12 cm x 3 cm and weighing approximately 1 kg.

From each granite sample, a specimen in the shape of a straight parallelepiped was extracted, measuring 12 cm × 3 cm x 2.0 cm (±0.005 cm) and weighing approximately 200 g. All faces of these specimens were sealed with four coats of sealing varnish, except for one of the larger faces, through which the ^{222}Rn is exhaled. After this procedure, the specimens were individually placed in 3.3 L glass containers, where they remained for 40 days to achieve secular equilibrium. Secular equilibrium is achieved when radionuclides belonging to the same radioactive series begin to exhibit the same activity concentration.

2.2. Measurement of ^{222}Rn activity concentration with the AlphaGUARD detector

After 40 days, the containers containing the granite specimens were opened and quickly transferred to identical, empty containers that had previously been sealed. This procedure was adopted to meet the boundary conditions of the activity concentration determination method, the one-dimensional long-term model, which requires that the atmosphere (container) be initially free of radon gas and that the specimen be saturated with this radionuclide, being the only source of gas exhalation in the system (container/specimen).

After transfer to the new container, the specimens were positioned longitudinally, with the larger side facing up. The portable AlphaGUARD detector (SAPHYMO GmbH) was used to measure the gas exhalation rate (^{222}Rn). The study was conducted during the second half of 2024.

The portable AlphaGUARD detector is a robust and accurate instrument widely used for the continuous, real-time measurement of ^{222}Rn activity concentration. It operates with a pulsed ionization chamber, which allows the detection of alpha particles emitted by ^{222}Rn and its decay products. The detector's ionization chamber has a useable volume of 0.62 L and operates over a measurement range from 2 to 2,000,000 Bq/m^3, with an instrumental calibration error of 3 % and a sensitivity of 1 cpm at 20 Bq/m^3 (SAPHYMO GmbH, 2023).

The AlphaGUARD equipment is calibrated through intercomparison work in CNEN laboratories by measuring ^{222}Rn from a closed-loop ^{226}Ra source, ensuring traceability and reliability. In addition to measuring ^{222}Rn activity concentration, this equipment is capable of simultaneously measuring ambient temperature, relative humidity, and atmospheric pressure through its integrated sensors. Fig. 1 shows the data acquisition system configuration.

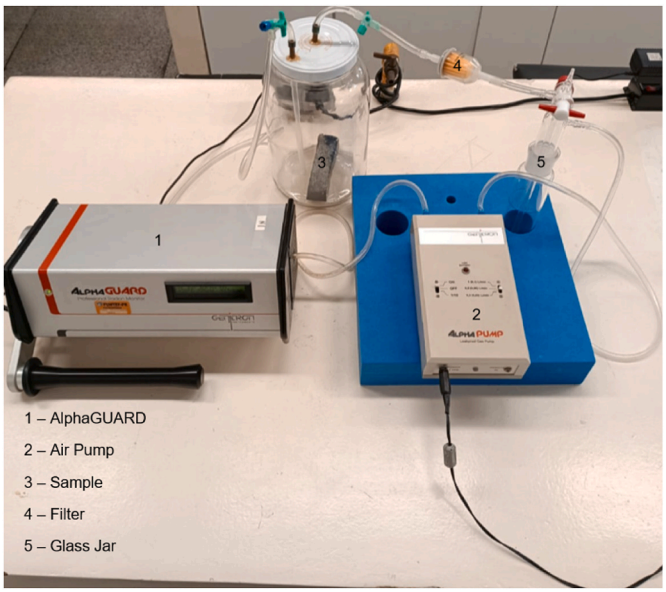

Fig. 1. – Data acquisition system.
Source: Own authorship

The AlphaGUARD was activated in 10 min/flow mode (one measurement every 10 min), and the air pump operated at a flow rate of 0.5 L/min. Each ^{222}Rn activity concentration measurement was performed for an uninterrupted period of 12 days, as required by the one-dimensional long-term model. This model determines that radon gas is exhaled by only one surface of the material, while the other surfaces are sealed.

2.3. Simplified semi-empirical physical-mathematical model

From the experimental data obtained by AlphaGUARD, the emanation coefficient (ω) of ^{222}Rn was determined using the simplified semi-empirical physical-mathematical model (Equation (1)), derived from the original model developed at the Laboratory of Applied Nuclear Physics of UTFPR (LFNA) by Barreto et al. (2017) (Equation (2)). The emanation coefficient of ^{222}Rn is directly proportional to the activity density of ^{226}Ra; thus, by calculating this parameter, it was possible to infer the activity concentration of ^{226}Ra. The fitting of the experimental data was performed using the gnuplot 5.4 program for the Indian Black, Fortaleza, Castle Gray, and Brazilian Gold granite samples.

$$f(t) = A + Be^{-\lambda t} + Ce^{-\lambda_1 t} \quad (1)$$

In equation (1), parameters A, B, C, and λ_1 are adjustable, while λ is the decay constant of ^{222}Rn, expressed in decays per minute.

- $A = \beta^2\omega$;
- $B = \beta^4\gamma\omega$;
- $\lambda = \frac{ln\ 2}{t_{1/2}}$;
- $\beta = \sqrt{\frac{D_I}{D_E}}$, • where D_I and D_E are the diffusion coefficients in the solid and in the external atmosphere;
- $\gamma = \frac{V_E}{V_I}$, with V_E and V_I being the volumes of the outer atmosphere and the solid, respectively.

$$\overline{n}_B(t) = \beta^2\frac{\omega}{\lambda}\left(1 - \gamma\beta^2 e^{-\lambda t}\right) - \overline{a}_1 e^{-\lambda_1 t} - \overline{a}_2 e^{-\lambda_2 t} \quad (2)$$

Table 1 presents the values of the ^{226}Ra activity concentration together with the errors associated with the measurements.

Initially, the specific activity of radium was obtained in Becquerel per cubic meter (Bq/m^3); later, it was divided by the density of granite, obtaining it in Becquerel per kg (Bq/kg).

Table 1
– Activity concentration of ^{226}Ra.

Sample	Activity concentration of ^{226}Ra (Bq/kg)
Indian Black	44,4 ± 7,2
Fortaleza	23,4 ± 8,0
Castle Gray	24,6 ± 7,8
Brazilian Gold	15,0 ± 5,0

Source: Own authorship

2.4. Gamma spectrometry with the HPGe detector

Gamma spectrometry with the HPGe detector was performed on samples of Indian Black, Fortaleza, Castle Gray, and Brazilian Gold granite. Eight hundred grams of the samples were collected and ground in a pan mill belonging to the laboratory of the Academic Department of Civil Construction (DACOC) at UTFPR, Curitiba Campus, for 180 s. This process resulted in an ultrafine powder.

The samples were sent to the Environmental Radiometry Laboratory of the Radiation Metrology Center of the Institute for Energy and Nuclear Research (IPEN/CNEN), located at Cidade Universitária, São Paulo, SP.

At IPEN, the materials were transferred separately to 100 mL Marinelli beakers and prepared for measurement. The gamma spectrometer used in the measurements is a 3″ coaxial Canberra XtRa, with extended energy for gamma radiation detection in the 3 keV to >10 MeV range. It features a carbon composite window and a solid-state hyperpure germanium (HPGe) semiconductor detector protected by a lead shield. This detector has a relative efficiency of 40 % and high resolution, allowing it to detect even small amounts of radionuclides present in a sample (Canberra. Germanium Detectors, 2019).

The HPGe detector works based on the principle of electron-hole pair formation when gamma radiation interacts with the semiconductor material. The gamma radiation energy is converted into a proportional electrical signal, which is then amplified and analyzed to determine the energy and intensity of the gamma rays emitted by the radionuclides present in the sample. The system's energy calibration is performed using standard radioactive sources with known gamma energies, such as ^{133}Ba, ^{152}Eu, ^{60}Co, while the efficiency calibration is performed with sources of similar geometry and matrix composition to the samples, ensuring accuracy in determining activity concentrations. The counting interval for each sample was 24 to ensure adequate counting statistics and reduce uncertainties.

For counting, the ground samples were hermetically sealed in Marinelli beakers and kept for a minimum of 40 days before measurement. This period is crucial for ^{226}Ra and its short-lived decay products (including ^{222}Rn) to reach radioactive equilibrium, ensuring that the measured ^{222}Rn activity is representative of the ^{226}Ra activity in the sample. The seal prevents the escape of gaseous ^{222}Rn, ensuring that all radon produced remains in the container and reaches equilibrium with its precursor.

Standard error calculations were performed using uncertainty propagation, considering the statistical uncertainties of the counts, detector calibration uncertainties, and uncertainties associated with sample preparation. The activity concentration of ^{226}Ra was determined indirectly from the intensity of the gamma peaks of its short-lived decay products (^{214}Pb and ^{214}Bi), assuming secular equilibrium. ^{228}Ra concentrations were determined from the gamma peaks of ^{228}Ac, while ^{238}U was inferred from ^{210}Pb, also in secular equilibrium. ^{40}K was measured directly by its characteristic gamma peak at 1460.8 keV. The Minimum Detectable Activity (MDA) for each radionuclide was calculated based on the Currie (1968) criteria, considering the detector background and counting time, ensuring that only statistically significant activities were reported.

Table 2 presents the activity concentrations of radionuclides ^{226}Ra, ^{228}Ra, ^{238}U and ^{40}K.

The activity of ^{238}U was determined from the measurement of ^{210}Pb, since these radionuclides are in secular equilibrium.

3. Results and discussion

From the experimental data regarding the ^{222}Rn concentration in the samples, obtained through the AlphaGUARD detector, together with the values of the effective sample volume (V_I), the container/external atmosphere volume (V_E), the effective sample length (L_I), the container/external atmosphere length (L_E), the exposed area of the specimen (S), and the radon decay constant (λ), the simplified semi-empirical physical-mathematical model (Equation (1)) was applied. Through this approach, the activity concentration of ^{226}Ra was inferred.

Fig. 2 presents graphs (a), (b), (c), and (d), adjusted for the mentioned samples, based on the application of Equation (1).

The figures show that the semi-empirical physical-mathematical model satisfactorily fits the experimental data obtained by the AlphaGUARD detector.

Based on the data presented in Tables 1 and 2, the difference between the 226Ra activity concentrations obtained by the different techniques employed was determined. Furthermore, using Equation (3), the difference error was calculated, defined as the difference between the estimated errors for each of the techniques used.

$$\sigma_{difference} = \sqrt{(\sigma_1)^2 + (\sigma_2)^2} \quad (3)$$

Where σ_1 and σ_2 represent the errors associated with each technique employed, these values being estimated based on the uncertainties inherent in the measurement and analysis processes used. The corresponding values are presented in Table 3.

The application of the normalized difference test (Z-score) to the ^{226}Ra activity concentration values obtained by the different techniques, as shown in Table 3, revealed that the values are statistically equal with 95 % certainty for the Indian Black, Fortaleza, Castle Gray, and Brazilian Gold samples.

The analysis of the results demonstrates the effectiveness of the simplified semi-empirical physical-mathematical model (Equation (1)) in estimating the ^{226}Ra activity concentration in granite samples. The statistical agreement with the results obtained by gamma spectrometry, a more established and expensive technique, validates the applicability of the proposed method. This validation is crucial, as it offers a more accessible alternative for assessing the radiological risk of construction materials, especially in regions with limited resources for complex laboratory analyses.

The ^{226}Ra activity concentrations varied among the samples, reflecting the natural heterogeneity of the granites' geological composition. The Indian Black and Fortaleza samples showed higher ^{226}Ra concentrations compared to Castle Gray and Brazilian Gold. These variations can be attributed to the different geographic origins and mineralogical compositions of the granites. Granites are intrusive igneous rocks formed by the slow cooling of magma. Their mineralogical composition (mainly quartz, feldspar, and mica) and, consequently, the content of radioactive elements, depend on the magma source and subsequent geological processes. For example, granites derived from more differentiated magmas or those that underwent enrichment processes in incompatible elements tend to have higher concentrations of U and Th, and, by extension, ^{226}Ra (UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, 2008).

Similar studies in the literature corroborate the variability of radionuclide concentrations in building materials and soils. Gümbür (2024) found different levels of radium and radon in bottled mineral waters, indicating that geological origin directly influences the presence of these elements. Similarly, Küçükönder et al. (2023) observed variations in natural radioactivity in soil samples from Kahramanmaraş, Turkey, attributing them to local geological characteristics.

The correlation between measured radon gas and radium is a fundamental aspect of radiological risk assessment. ^{222}Rn is a decay product of ^{226}Ra, and its concentration in indoor environments is directly influenced by the amount of ^{226}Ra present in building materials. Radon exhalation from granite samples, measured by AlphaGUARD, reflects ^{226}Ra activity, especially when secular equilibrium is reached. Analyzing this correlation is crucial for predicting radon exposure in environments where these granites are used. To contextualize the radiological impact, the ^{222}Rn concentration in a standard room (volume = 40 m^3) was estimated from the measured ^{226}Ra-specific activities. Considering a mass of 100 kg of Indian Black granite and a realistic emanation factor of 0.2 (20 %), the ^{222}Rn airborne concentration (CRn) would be approximately 22.2 Bq/m^3. This value corresponds to an annual effective dose of approximately 0.56 mSv/year, calculated using the parameters in Equation (4).

The annual effective dose (AED) due to inhalation of ^{222}Rn and its progenitors in indoor environments was calculated using Equation (4) (ICRP - International Commission on Radiological Protection, 2010).

$$DEA = C_{Rn}\ x\ F_{eq}\ x\ T_{ocup}\ x\ D_{conv} \quad (4)$$

Where:

- C_{Rn} is the average activity concentration of ^{222}Rn in the air (Bq/m^3).
- F_{eq} is the equilibrium factor between ^{222}Rn and its decay products (usually 0.4 for indoor environments).
- T_{ocup} is the annual occupancy time (usually 7000 h/year for residential buildings).
- D_{conv} is the dose conversion coefficient (usually 9 nSv/(Bq h m^{-3}) for ^{222}Rn).

The estimated annual doses for all samples were well below the reference value of 2.4 mSv/year, which represents the global average natural radiation exposure according to UNSCEAR. This indicates that, even for the most active granite, the radiological risk associated with its use as a finishing material in well-ventilated environments is low. However, scenarios with large exposed surfaces or poor ventilation could result in higher doses, justifying specific monitoring.

The results of this study have important implications for the construction industry and regulatory agencies. Validating a more accessible method for determining ^{226}Ra concentration allows for more widespread monitoring. However, it is crucial to recognize the limitations of the model: it is a simplification and relies on the assumption of secular equilibrium between ^{226}Ra and ^{222}Rn, which may not be fully achieved. Future studies could explore the influence of environmental factors (such as humidity and pressure) on the radon exhalation rate.

Table 2
– Concentration of activity of ^{226}Ra, ^{228}Ra, ^{238}U and ^{40}K.

Sample	Concentration of activity (Bq/kg)			
^{226}Ra	^{228}Ra	^{238}U	^{40}K	
Indian Black	70,7 ± 7,3	49 ± 14	46 ± 12	1067 ± 72
Fortaleza	41,6 ± 4,3	44 ± 11	54,9 ± 6,8	1273 ± 84
Castle Gray	37,3 ± 3,9	117 ± 30	29,0 ± 4,5	1442 ± 95
Brazilian Gold	23,9 ± 2,7	57 ± 15	–	1091 ± 73

Source: Own authorship

4. Conclusion

The semi-empirical physical-mathematical model proved to be an economically viable technique for determining the ^{226}Ra activity concentration in construction materials when compared to gamma spectrometry, whose potential application is widely recognized. Application of the model requires only that the ^{222}Rn gas concentration be measured

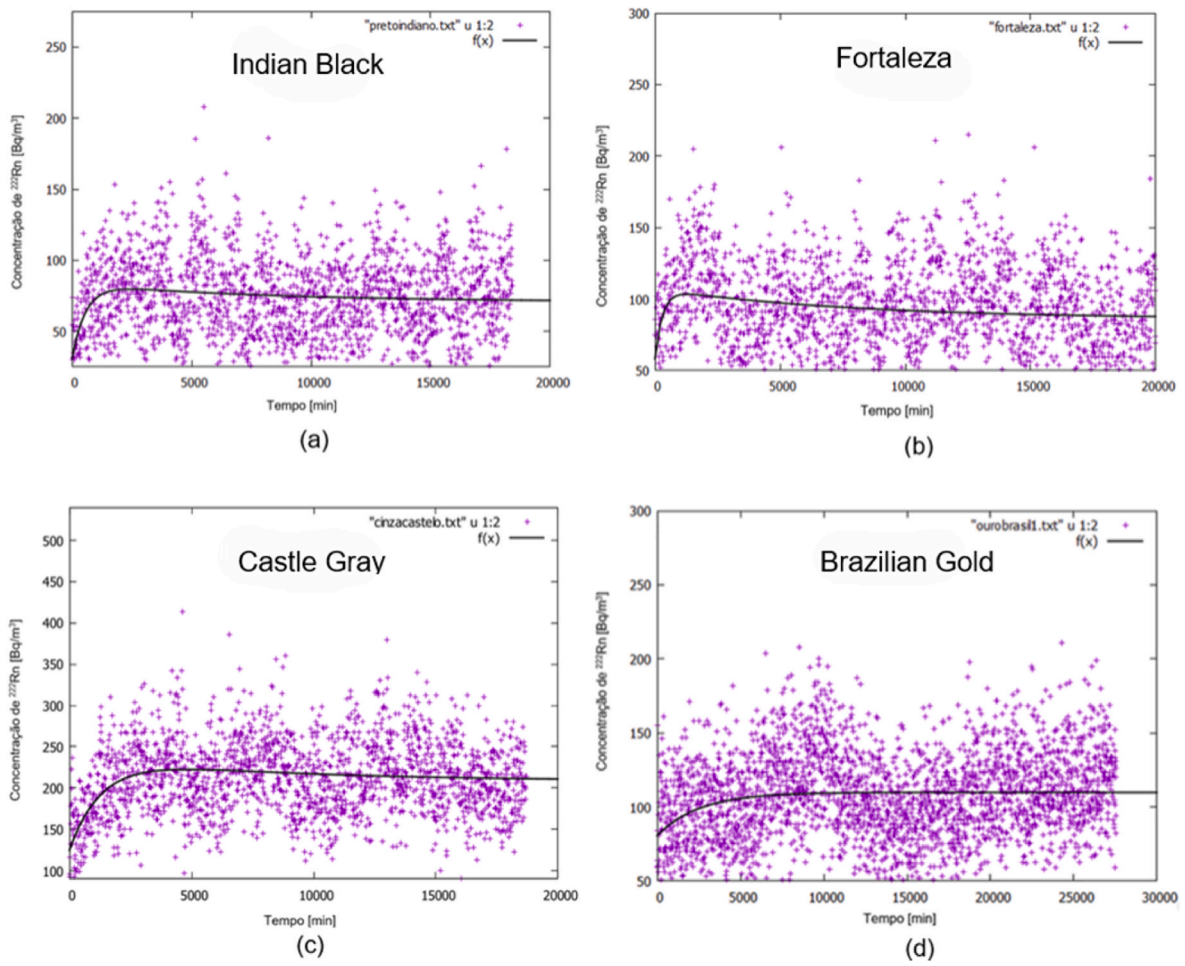

Fig. 2. – Curves adjusted by the physical-mathematical model.
Source: Own authorship

Table 3
– Difference between ^{226}Ra activity concentrations and difference in errors.

Applied Technique				
Sample	Simplified model	Gamma spectrometry	Difference in activity (Bq/kg)	Difference error (Bq/kg)
Specific activity of ^{226}Ra (Bq/kg)				
Indian Black	44,4 ± 7,2	70,7 ± 7,3	26,3	±10,2
Fortaleza	23,4 ± 8,0	41,6 ± 4,3	18,2	±9,0
Castle Gray	24,6 ± 7,8	37,3 ± 3,9	12,7	±8,7
Brazilian Gold	15,0 ± 5,1	23,9 ± 2,7	8,9	±5,7

Source: Own authorship

by the AlphaGUARD detector with the sample in a closed atmosphere. The data presented in Table 3 show that the values obtained for the ^{226}Ra activity concentration using the different techniques present a statistical agreement of 95 %, suggesting the potential application of the physical-mathematical model. Furthermore, the values obtained for the ^{226}Ra activity concentration are below the limit of 1000 Bq/kg, established by national and international agencies, such as CNEN and IAEA, respectively, demonstrating that there are no radiological restrictions regarding the use of the analyzed granites as construction materials.

CRediT authorship contribution statement

Ricardo Rossasi Geraldo: Writing – original draft. **Sergei Anatolyevich Paschuk:** Supervision. **Janine Nicolosi Correa:** Supervision. **Valeriy Denyak:** Supervision. **Monique Soares de Oliveira:** Writing – original draft. **Guilherme Soares Zahn:** Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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