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Determination of natural radioactivity levels in drinking water and soil at APA Passaúna (Curitiba, PR)

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Abstract: Natural radioactive materials have been part of the ecosystem since the formation of the Earth. Monitoring these nuclides in rocks, soils, groundwater, and air is essential for the scientific community and for implementing mitigation measures. This is because radon decay products are the second leading global cause of lung cancer, after tobacco. This study focused on conducting a radiometric survey of properties in the APA Passaúna, Curitiba (PR). The AlphaGUARD PQ2000 PRO equipment was used to measure the concentrations of ^{222}Rn and ^{226}Ra in well water and ^{222}Rn and ^{220}Rn in soil air. Gamma spectrometry was conducted with the portable scintillator BRD-AT6101C. The analysis revealed the presence of ^{40}K in all the investigated sites, as expected, along with lower amounts of ^{226}Ra and ^{232}Th. Radiation dose equivalents were generally low, except at a specific point where the estimated dose was 1.65 μSv/h, with a ^{220}Rn concentration of (664 ± 36) kBq/m³. Seven out of the nine wells studied showed a committed effective dose value for the ingestion and inhalation of drinking water above the reference level set by the WHO (0.1 mSv/year). The excess values ranged from 0.106 to 0.385 mSv/year. In contrast, radon concentrations in indoor air ranged from (30 ± 2) to (270 ± 20) Bq/m³, within normal parameters. According to the CNEN standard NN 3.01, the study complies with Brazilian radioprotection standards and has significant social relevance, especially in a region where water is predominantly sourced from wells. These data are preliminary for the southern region of Brazil and highlight the need for further scientific research.

Keywords: natural radioactivity, radon, groundwater, gamma spectrometry.

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Determinação dos níveis de radioatividade natural em águas potáveis e no solo na APA Passaúna (Curitiba, PR)

Resumo: Os materiais radioativos naturais têm sido parte do ecossistema desde a formação da Terra. O monitoramento desses nuclídeos em rochas, solos, águas subterrâneas e no ar é essencial para a comunidade científica e para a implementação de medidas de mitigação. Isso se deve ao fato de que os produtos do decaimento do radônio são a segunda principal causa global de câncer de pulmão, atrás apenas do tabaco. Este estudo concentrou-se na realização de um levantamento radiométrico em propriedades situadas na APA do Rio Passaúna, em Curitiba (PR). Utilizou-se o equipamento AlphaGUARD PQ2000 PRO para medir as concentrações de ^{222}Rn e ^{226}Ra nas águas dos poços e de ^{222}Rn e ^{220}Rn no ar do solo. A espectrometria gama foi conduzida com o cintilador portátil BRD-AT6101C. A análise revelou a presença de ^{40}K em todos os terrenos investigados, conforme esperado, além de menores quantidades de ^{226}Ra e ^{232}Th. As doses equivalentes de radiação foram, em geral, baixas, exceto em um ponto específico onde a dose estimada foi de 1,65 µSv/h, com uma concentração de ^{220}Rn de (664 ± 36) kBq/m³. Sete dos nove poços estudados apresentaram valor de dose efetiva comprometida para ingestão e inalação de água potável acima da referência estipulada pela OMS (0,1 mSv/ano). Os valores excedentes variaram de 0,106 a 0,385 mSv/ano. Em contraste, as concentrações de radônio no ar das habitações variaram de (30 ± 2) a (270 ± 20) Bq/m³, dentro dos parâmetros normais. O estudo está em conformidade com as normas de radioproteção brasileiras, conforme a norma NN 3.01 da CNEN, e possui relevância social significativa, especialmente em uma região onde a água é predominantemente obtida de poços. Estes dados são preliminares para a região sul do Brasil e destacam a necessidade de mais pesquisas científicas.

Palavras-chave: radioatividade natural; radônio; águas subterrâneas; espectrometria gama.

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1. INTRODUCTION

Radon is a radioactive gas from the decay of naturally active primordial isotopes in the Earth's crust. Generally, exposure to this gas in well-ventilated environments does not pose a life-threatening risk. However, research conducted across various continents indicates that inhalation of radon isotopes in enclosed spaces causes significant harm to human health [1, 2, 3].

In the 1980s, the International Agency for Research on Cancer (IARC) classified radon as a human carcinogen, placing it in the same group as asbestos and benzene [4].

Recently, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) reported that radon is the leading cause of lung cancer among non-smokers and the second leading cause among smokers [5]. Consequently, the issue of radon inhalation and its short-lived progeny has become a public health concern, warranting research that monitors the activity of this gas in residential environments.

Radon activity originates from the emanation of radon present in the soil and rocks into the air within dwellings. Another significant source of exposure to ^{222}Rn is groundwater. Direct ingestion of water containing dissolved radon leads to the deposition of its progeny in internal organs. The successive decays of ^{222}Rn products release energy quanta or α and β particles that damage human tissues and increase the risk of cancer development [6, 7].

However, approximately 90% of the dose attributable to radon dissolved in drinking water comes from inhaling this gas when it is released during agitation or heating of the water. For this reason, routine activities such as bathing and dishwashing can pose health risks if exposure occurs in a prolonged and constant manner [7, 8].

Besides radon, another significant isotope contributing to human exposure is ^{40}K, which is widely disseminated in the Earth's crust within specific sedimentary coverings [9].

Therefore, monitoring active nuclides in rocks, soils, drinking water, and air in living environments is of paramount importance, both to provide data to the scientific community and to enable mitigative interventions for stakeholders. For that reason, the objective of this study was to conduct a radiometric survey in private properties located in the Rio Passaúna Environmental Protection Area (Curitiba, PR).

1.1. Passaúna Environmental Protection Area

The Passaúna River Environmental Protection Area, known as the APA Passaúna, was established by State Decree Nº. 458 in 1991 and encompasses parts of the municipalities of Curitiba, Campo Largo, Araucária, and Almirante Tamandaré [10]. The area was created to preserve and restore the Passaúna River watershed, which houses the water source that supplies approximately 20% of the drinking water consumed by Curitiba and its metropolitan region. A management plan was developed to impose restrictions on land and water use in the region to achieve this goal. Those restrictions, combined with the local community's economic and cultural practices, have resulted in the predominant consumption of water from artesian and semi-artesian wells by the residents.

Additionally, preliminary gamma spectrometry measures have identified locations within the area with relatively elevated levels of ^{232}Th and ^{226}Ra, further justifying the selection of this area for the development of research [11].

2. MATERIALS AND METHODS

This study was conducted on eight private properties in the Passaúna Environmental Protection Area.

Water samples were collected from nine wells on all characterized properties, and surface gamma spectrometry of the soil was performed. These preliminary measurements

indicated the need to monitor alpha activity in the soil air at specific points and evaluate the indoor air of the residences.

2.1. Low-Resolution Gamma Spectrometry

Gamma spectrometric data were obtained using the portable AT6101C detector (Atomtex), which has two data acquisition units: the BDKG-11M for gamma radiation detection and the BDKN-05 for neutron detection. Gamma spectrometry uses a sodium iodide scintillation crystal doped with thallium, with dimensions of 63x63 mm, coupled with a photomultiplier system and a pulse discrimination and analysis circuit (factory calibration – 2018).

Figure 1 shows the data acquisition units for (a) gamma and (b) neutrons, (c) the electronic microcontroller, and (d) the monitor controlling the measurements.

Figure 1: Portable detector AT6101C (Atomtex).

Source: Authors (2024).

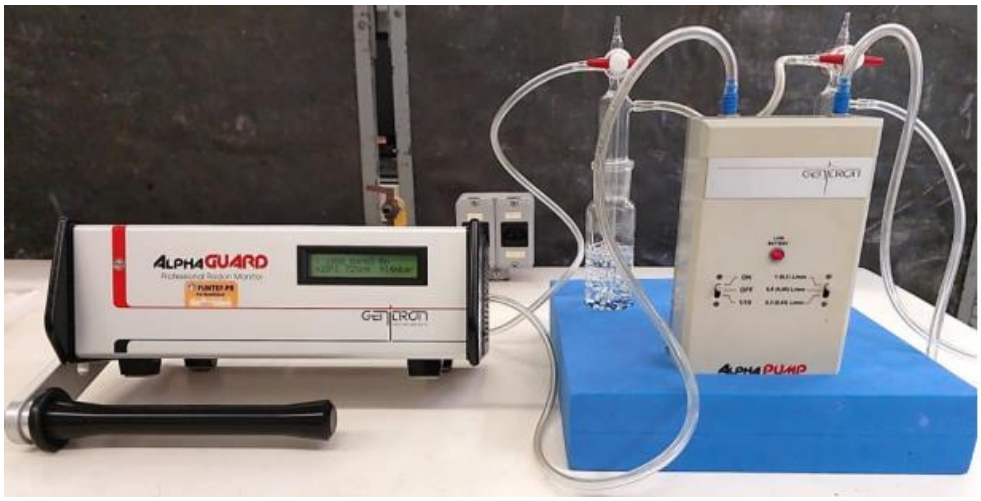
Using the walkover method, the entire surface area of each property was scanned (about tens of thousands of square meters).

2.2. Activity of ^{222}Rn and ^{226}Ra in Well Water

The water samples collected from the residents' wells were evaluated using the AlphaGUARD PQ2000 PRO equipment (Saphymo, now Bertin Technologies) coupled with the AquaKIT in a closed circuit, as shown in Fig. 2. According to the equipment manual, the detection efficiency ranges from 2 to 2M Bq/m³ (0.002 to 2000 Bq/L).

Following the manufacturer's guidelines, each measurement was performed with 100 ml of water. A total of 1 liter was collected per well for further examination.

Figure 2: Experimental setup for drinking water analyzes (AlphaGUARD PQ2000 PRO).



Source: Authors (2024).

Five measurements of ^{222}Rn concentration were performed on each water sample, with intervals of 4 days between each measurement, approximately equal to the element's half-life. Finally, an additional measurement was taken 38 days after sample collection to measure the ^{222}Rn concentration in secular equilibrium with ^{226}Ra. Subsequently, concentration versus time graphs were constructed and fitted using the following equation:

$$A_{222}(t) = A_{0_{226}}\left(1 - e^{-\lambda_{222}t}\right) + A_{0_{222}}\left(e^{-\lambda_{222}t}\right)$$

The first term is related to the ^{222}Rn produced by the decay of ^{226}Ra dissolved in the water. In contrast, the second term is related to the ^{222}Rn produced by the decay of ^{226}Ra located in another compartment but released into this water.

2.3. Activity of ^{222}Rn and ^{220}Rn in Soil Air

In the soil, the measurement of radon and thoron isotopes was performed using the AlphaGUARD PQ2000 PRO equipment (Saphymo, now Bertin Technologies), but in an open circuit and coupled with the capillary probe, as shown in Fig. 3.

Figure 3: Experimental setup for soil gas analyzes (AlphaGUARD PQ2000 PRO).

Source: Authors (2024).

The soil air measurement points were selected where gamma spectrometry identified high concentrations of Th and Ra isotopes.

Initially, the suction pump was activated and remained on for 10 minutes to allow mixing the soil air with the air in the cavity. The measurement was conducted for an additional 10 minutes under a steady airflow. After this period, the measurement continued with the pump turned off for 10 minutes, aiming to observe thoron decay.

2.4. The activity of ^{222}Rn Indoors

Solid-state detectors were installed in all rooms of two properties to monitor the indoor environments of the residences.

Urban and Piesch (1981) proposed the diffusion chamber for the measurement process, with a semi-spherical shape and a sensitive volume of 7.1 cm³. This configuration helps reduce the deposition of Rn progeny on the radiosensitive material. Additionally, fiberglass filters were used at the air inlet of the chamber to filter particulates and allow the permeability of ^{222}Rn.

The assembly of the diffusion chambers with the filter and CR-39 is illustrated in Fig. 4. The filter protects the detector from damage and aerosols while the chamber homogenizes the detection process.

Figure 4: Difusion camara.

Source: Authors (2024).

For calculating the concentration of ^{222}Rn in the air, the calibration equation developed by Corrêa in collaboration with the Centro de Desenvolvimento da Tecnologia Nuclear (CDTN) and the National Institute of Radiological Sciences was used [12, 13].

$$C = (405 \pm 30)d \left[\frac{Bq}{m^3}\right]$$

Where 'd' represents the number of tracks per hour of exposure in 1 cm² of the detector.

The exposures were conducted over approximately four months in both residences. The development of the detectors followed the protocol established by Corrêa [12], involving immersion of the detectors in a 6.25 M NaOH solution for 14 hours at 70°C.

3. RESULTS AND DISCUSSIONS

The analysis of gamma spectrometric data was performed using the GARM® (Geolocation Application for Radiation Monitoring) software, which allows for overlaying the collected dose rate data at one meter above the surface (DR1) with Google Maps® imagery layers. The evaluation of radon concentration data in water and soil air was conducted using DataEXPERT® software. Data processing and constructing exponential graphs were carried out using OriginLab® software.

Table 1 presents the obtained values of ^{222}Rn and ^{226}Ra concentrations dissolved in the well (W1A-W8) water studied at each property.

Table 1: Compiled results of drinking water analyzes.

Well	Concentration of ^{222}Rn (Bq/l)	Concentration of ^{226}Ra (Bq/l)
W1A	38.2 ± 2.4	0.05 ± 0.15
W1B	3.0 ± 0.1	0.09 ± 0.05
W2	18.4 ± 1.4	0.20 ± 0.28
W3	17.32 ± 0.82	0.06 ± 0.16
W4	32.7 ± 1.8	0.26 ± 0.35
W5	5.4 ± 0.2	0.19 ± 0.09
W6	9.46 ± 0.49	0.16 ± 0.19
W7	2.640 ± 0.058	0.391 ± 0.021
W8	3.114 ± 0.071	0.503 ± 0.024

According to the regulatory position 3.01/012 of CNEN, the specific activities of the two radionuclides (^{222}Rn and ^{226}Ra) were accounted for separately.

The committed effective dose for one year (CED) was calculated for each water sample using the equation below [14].

$$D_{ing} = \Sigma_j \, TI_{água} * C^{j}_{água} * FCD^{j}_{ing}$$

The parameter $Ding$ represents the committed effective dose due to the ingestion of contaminated water, expressed in Sv/year; the parameter $TIágua$ accounts for the amount of water ingested by an individual over the year, defined by WHO as 730 liters/year for adults, equivalent to daily consumption of 2 liters; $Cágua\ j$ is the concentration of radionuclide j dissolved in the water sample, in Bq/L; $FCDing\ j$ is the dose conversion factor for the ingestion of radionuclide j, in Sv/Bq.

The results of the dose calculations from the ingestion of ^{222}Rn and ^{226}Ra isotopes are organized in Table 2.

Table 2: Committed effective dose (CED) for ingestion of ^{222}Rn and ^{226}Ra

Well	CED ing ^{222}Rn (mSv/year)	CED ing ^{226}Ra (mSv/year)	CED ing total (mSv/year)
W1A	0.278860	0.010220	0.289080
W1B	0.021900	0.018396	0.040296
W2	0.134612	0.040880	0.175492
W3	0.126436	0.012264	0.138700
W4	0.238418	0.053144	0.291562
W5	0.039566	0.038836	0.078402
W6	0.069058	0.032704	0.101762
W7	0.019272	0.079920	0.099192
W8	0.022732	0.102813	0.125545

All wells showed that the combined CED values for the ingestion of ^{222}Rn and ^{226}Ra exceed the [15] guidance level of 0.02 mSv/year for a member of the general public.

However, approximately 90% of the effective dose attributable to radon in drinking water comes from inhalation of the gas, which is released during the agitation or heating of contaminated water during routine household activities. Therefore, the annual CED for inhalation of ^{222}Rn dissolved in the well water was calculated using the methodology described in the [15] publication, as follows:

$$D_{inal} = C_{Rn_{água}} * R * F_{eq} * I * FCD_{inal}^{Rn}$$

Where the parameter $Dinal$ represents the committed effective dose due to the inhalation of water contaminated with ^{222}Rn, expressed in μSv/year; the parameter $CRnágua$ accounts for the concentration of ^{222}Rn dissolved in the water sample [kBq/m^3]; R represents the increment coefficient of ^{222}Rn in indoor air from the concentration of ^{222}Rn dissolved in the water (10^{-4}); Feq is the equilibrium factor between ^{222}Rn and its progeny (0.4); I accounts for the average indoor occupation time by a typical individual (7000 hours/year); $FCDinal\ Rn$ is the dose conversion factor for the inhalation of ^{222}Rn [(9 nSv (Bq hm^{-3})$^{-1}$)]. The results of the dosimetric calculations are presented in Table 3.

Table 3: Committed effective dose (CED) for inhalation of ^{222}Rn

Well	CED inh ^{222}Rn (mSv/year)
W1A	0.096264
W1B	0.007560
W2	0.046469
W3	0.043646
W4	0.082303
W5	0.013658
W6	0.023839
W7	0.006653
W8	0.007847

By summing the CEDs for ingestion and inhalation of ^{222}Rn and ^{226}Ra, quantities above the [16] reference level of 0.1 mSv/year were obtained for wells W1A, W2, W3, W4, W6, W7, and W8, as shown in Table 4.

Table 4: Final result of drinking water analyzes.

Well	CED total (mSv/year)
W1A	0.385
W1B	0.048
W2	0.222
W3	0.182
W4	0.374
W5	0.092
W6	0.125
W7	0.106
W8	0.133

It is scientifically agreed that local geology significantly influences the concentration of radon in ecosystem compartments [17, 18, 19]. However, the activity values of ^{222}Rn and ^{226}Ra in the groundwater obtained in this study are significant.

The minimum and maximum DR1 values found at each property are presented in Table 5. The detected isotopes at the surface were also compiled.

Table 5: Collected values from surface gamma spectrometry.

Property	Identified Nuclides	DR1 min (µSv/h)	DR1 max (µSv/h)
P1	^{40}K, ^{226}Ra, ^{232}Th, ^{228}Th	0.030	1.650
P2	^{40}K	0.043	0.087
P3	^{40}K, ^{226}Ra, ^{232}Th	0.072	0.210
P4	^{40}K	0.041	0.087
P5	^{40}K	0.065	0.104
P6	^{40}K	0.037	0.102
P7	^{40}K, ^{232}Th	0.019	0.159
P8	^{40}K, ^{226}Ra, ^{232}Th	0.027	0.166

The gamma spectrometric data indicated relatively low values. The exception was a point with a DR1 of 1.65 μSv/h located at Property 1. Alpha activity measurements of the soil air showed the predominance of ^{220}Rn, consistent with the surface gamma spectrometry that indicated the presence of ^{232}Th and ^{228}Th at this location.

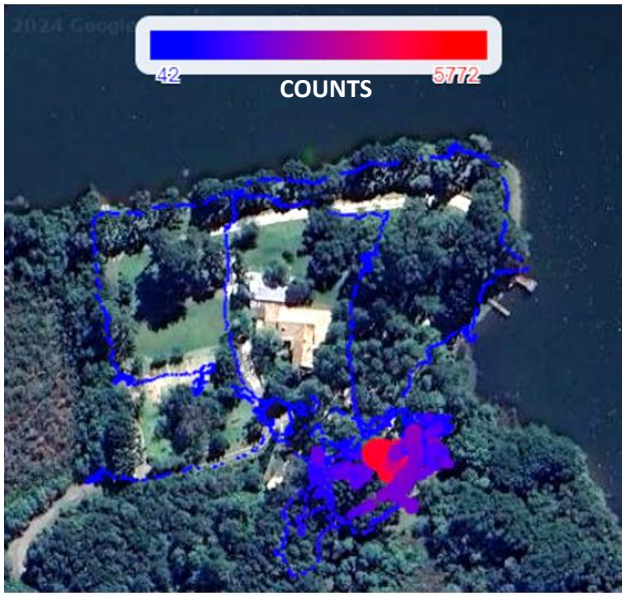
The significant detection of ^{40}K across all the terrains is consistent with the geological and mineral resource map of the state of Paraná, compiled by Besser et al. [20], which places the APA Passaúna over a granite/gneiss rock formation. This structure may contain potassium feldspar, typically rich in ^{40}K. Areas with sedimentary cover containing arkosic rocks may accumulate ^{40}K in certain regions. Additionally, the igneous origin of granite/gneiss may lead to the presence of uranium and its decay products, justifying the identification of ^{226}Ra and ^{214}Bi. It is important to note that radiation exposure from rocks is low, as the system is closed. However, weathering-induced changes in these rocks can significantly increase exposure.

The Google imagery layers generated within the GARM® software are outdated and may not accurately represent the studied terrains. Furthermore, the detector acquires spectra every 5 seconds (in the default configuration), generating various georeferenced data stored in .txt format. The software compiles this data to create markers on the maps that aggregate a certain amount of data, which results in the suppression of data acquired by the detector.

For this reason, a Python script was developed to generate more accurate maps encompassing all the acquired data. This script can read the .txt files generated by the spectrometry, select the columns of interest, and overlay the gamma spectrometric data onto the most recent Google Earth® imagery layer.

Figure 5 shows the image generated by the script for mapping Property 1. The columns of 'count', 'latitude', and 'longitude' were selected from the text file, forming ordered triplets representing each data acquisition point.

Figure 5: Map generated by python code (property 1) executed in the virtual environment of Google Colab®.



Source: Authors (2024).

The script uses the latitude and longitude data extracted from the text file to determine the exact location of each mapped point. The data processing involves excluding columns with irrelevant data and removing rows where latitude, longitude, or count is 0. Subsequently, a function is called to read the processed data [21].

In this script, the count values were normalized, and a function was defined to calculate the color of the markers based on the count in a color scale, both of which are optional actions. The legend can be customized according to the author's preferences.

Only properties 1 and 7 have solid-state detectors installed to measure indoor air. In the first, due to the high DR1 values obtained in gamma spectrometry. In the second, at the request of the residents.

Table 6 and Table 7 present the results of indoor radon measurements in dwellings 1A and 1B, respectively, both belonging to property 1.

Table 6: Indoor radon concentration in dwelling 1A.

Room	Concentration of ^{222}Rn (Bq/m³)	Error (Bq/m³)
Child's bedroom	79	6
Master bedroom	30	2
Living room-kitchen area	48	4

Table 7: Indoor radon concentration in dwelling 1B.

Room	Concentration of ^{222}Rn (Bq/m³)	Error (Bq/m³)
Master bedroom	48	4
Office	49	4
Storage room	89	6
Living room-kitchen area	62	5

Table 8 and Table 9 present the results of indoor radon measurements in dwellings 7A and 7B, respectively, both belonging to property 7.

Table 8: Indoor radon concentration in dwelling 7A.

Room	Concentration of ^{222}Rn (Bq/m³)	Error (Bq/m³)
Living room-kitchen area	66	5
Master bedroom	80	6
Bathroom	34	2

Table 9: Indoor radon concentration in dwelling 7B.

Room	Concentration of ^{222}Rn (Bq/m³)	Error (Bq/m³)
Master bedroom	270	20
Master bathroom	155	11
Child's bedroom	51	4
Kitchen	64	5
Living room	152	11
Bathroom	67	5
Garage	67	5

Radon-222 concentrations in indoor air ranged from (30 ± 2) to (270 ± 20) Bq/m³, in both dwellings. The highest values were obtained in the suite (270) and in the bathroom of the suite (155) of residence 7. Although the values are within the investigation level established by CNEN (between 100 and 300 Bq/m³), the highest value did not exceed the upper limit [14]. In addition, the residents reported traveling during this period, a fact that may influence these values.

The exposures were carried out between the months of April and July.

4. CONCLUSIONS

The radiometric survey conducted in this study included external surface gamma spectrometry, quantification of the specific activities of ^{222}Rn and ^{226}Ra dissolved in drinking water samples, and monitoring of indoor ^{222}Rn in private properties located within the APA do Passaúna.

The values found for the absorbed dose rate outdoors from natural gamma radiation at 1 meter from the surface were relatively low, ranging from 0.019 to 0.210 μSv/h. The exception was an anomalous point at Property 1, with a DR1 of 1.650 μSv/h. The specific treatment performed at this point involved evaluating the alpha activity of radon and thoron isotopes in the soil air and installing solid-state detectors in the surrounding residences. The study indicated the predominance of the ^{220}Rn isotope in the soil gas, with a concentration of (664 ± 36) kBq/m³. No significant increases in indoor radon were observed in the respective residences, as the anomalous point is not located directly beneath the buildings, and the predominant isotope has a short half-life.

The significant detection of ^{40}K is consistent with the local geology, where the rock formation consists of granite/gneiss with sedimentary cover points, which may contain potassium feldspar.

Regarding the well water analyses, it was observed that, in all cases, the amounts were below the international consensus intervention limit (100 Bq/m³). However, seven out of the nine wells studied had effective doses for ingestion and inhalation of drinking water above the reference level set by WHO (0.1 mSv/year). The excess values ranged from 0.106 to 0.385 mSv/year.

This work aligns with the federal concern regarding the basic radioprotection requirements in Brazilian territory, as outlined in the draft standard NN 3.01 from CNEN, published in March 2024.

This survey also contributes to the acquisition of radiometric data still scarce in the southern region of the country and has immediate social relevance, as it provides both information to the scientific community and the possibility of mitigation interventions for stakeholders, aiming to minimize the risk of cancer development among residents of the region.

In this regard, it is necessary to survey more properties where high radon concentrations in groundwater were found.

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CONFLICT OF INTEREST

The authors declare the following academic commitments, which may be considered potential competing interests: This report results from a master's dissertation presented to the Graduate Program in Electrical Engineering and Industrial Informatics (CPGEI) of the Federal University of Technology – Paraná – Brazil (UTFPR).

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