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Original Article

Committed Effective Dose by Ingestion Food Grown in PEPB

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Abstract: Radionuclides can be incorporated through ingestion and within the human body, they become a source of internal exposure. Therefore, identifying and quantifying the radionuclides present in food is an important step in environmental monitoring and radiological protection. From this perspective, the activity concentrations of ^{40}K, ^{226}Ra, ^{228}Ra and ^{228}Th were calculated for six organic crop samples: avocado, cabbage, carrot, persimmon, pineapple and onion, from the PEPB (stands for Pedra Branca State Park), Conservation Unit. Radionuclide analysis wasconducted using gamma spectrometry with the use of a High Purity Germanium (HPGe) detector in conjunction with the Genie 2000 software package. The activity concentrations of the radionuclides were calculated based on simulated detection efficiencies. The highest activity concentration of ^{40}K was obtained for the carrot (101.83 ± 15.47) Bq.kg$^{-1,}$ and the highest effective dose value, 1.64 µSv.year^{-1}, was obtained for avocado. The concentration of ^{226}Ra activity was below the minimum detectable activity in all samples. Concentrations of ^{228}Ra were only detected in cabbage and carrots. Activity concentrations of ^{228}Th were detected in all samples, although at low levels. Estimated ingestion doses are within the limits established by the ICRP 119. Thus, the consumption of these organic foods grown in the PEPB is radiologically safe.

Keywords: gamma spectrometry, committed effective dose, organic crop, Pedra Branca State Park.

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Dose Efetiva Comprometida por Ingestão de Alimentos Cultivados no Parque Estadual da Pedra Branca

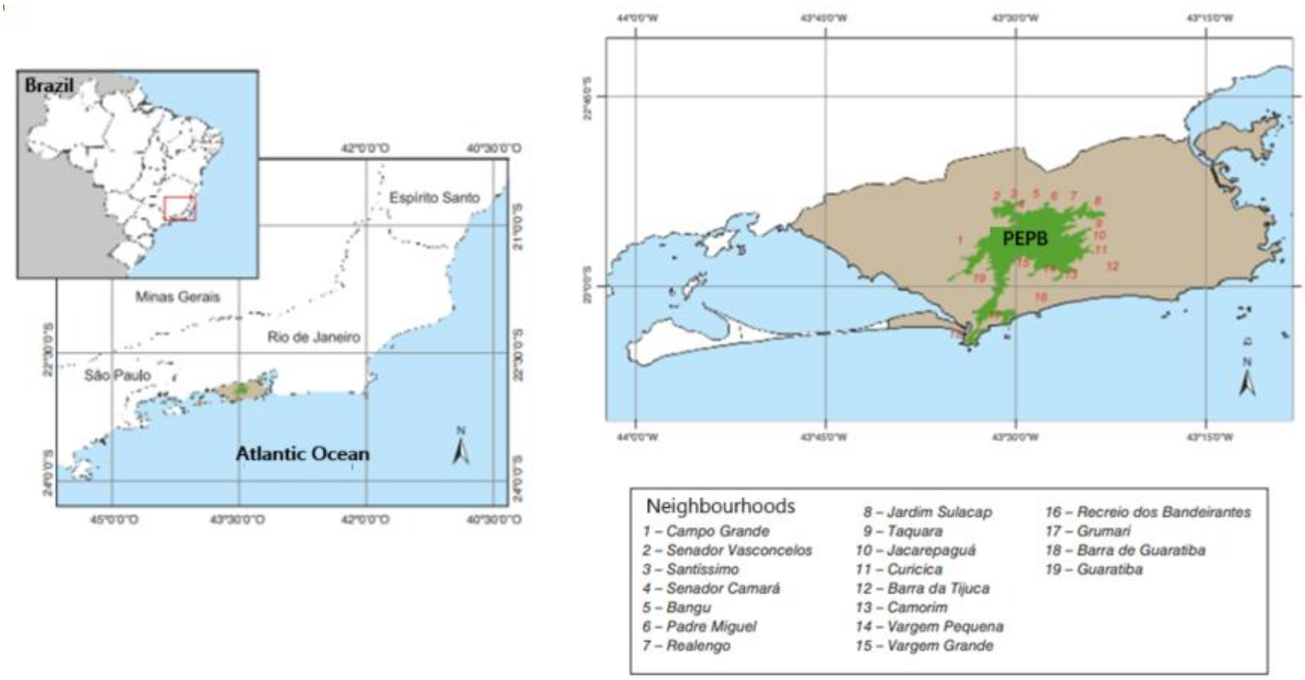
Resumo: Os radionuclídeos podem ser incorporados por meio da ingestão e no corpo humano, tornam-se uma fonte de exposição interna. Por conseguinte, identificar e quantificar os radionuclídeos presentes nos alimentos é um passo importante na monitorização ambiental e na proteção radiológica. Nesta perspectiva, as concentrações de atividade de ^{40}K, ^{226}Ra, ^{228}Ra e ^{228}Th foram calculadas para seis amostras de cultivo orgânico: abacate, couve, cenoura, caqui, abacaxi e cebola, do Parque Estadual da Pedra Branca (PEPB), Unidade de Conservação. A análise dos radionuclídeos foi realizada por espectrometria gama com o uso de um detetor de germânio de alta pureza (HPGe) em conjunto com o pacote de software Genie 2000. As concentrações de atividade dos radionuclídeos foram calculadas com base em eficiências de deteção simuladas. A concentração de atividade mais elevada de ^{40}K foi obtida para a cenoura (101.83 ± 15.47) Bq.kg^{-1} e o valor mais elevado de dose efetiva, 1.64 μSv.ano^{-1}, foi obtido para o abacate. A concentração de atividade do ^{226} Ra ficou abaixo da atividade mínima detectável para todas as amostras. Concentrações de atividade do ^{228} Ra foram encontradas somente na couve e na cenoura. Concentrações de atividade de ^{228}Th foram detectadas em todas as amostras, embora em baixos níveis. As doses estimadas por ingestão estão dentro dos limites estabelecidos pela ICRP 119. Portanto, o consumo desses alimentos orgânicos cultivados no PEPB é radiologicamente seguro.

Palavras-chave: espectrometria gama, dose efetiva comprometida, cultivo orgânico, Parque Estadual da Pedra Branca.

1. INTRODUCTION

PEPB is a Conservation Unit (UC) located in the municipality of Rio de Janeiro. Administered by the State Environment Institute (INEA), it covers an area of just over 12,000 hectares and encompasses 17 neighborhoods in the West Zone. The park is located between the geographical coordinates 22053'5" south latitude - 43034'36" west longitude - 2304'21" south latitude and 43022'46" west longitude [1].

Figure 1: PEPB highlighted on the maps of Rio de Janeiro state and municipality



Source: Modified from INEA(2013).

The site offers various types of activities, including hiking along its many trails, horseback riding, various observation points and diving in its waterfalls and rivers [1]. Its geological formation is made up of a group of rocks with different lithologies, involving granitoids and gneisses [2]. The presence of these rocks presupposes higher concentrations of natural radionuclides [3]. Despite UCs conceptually not allowing agricultural practices, however, at the time of the park's creation, family farmers already living in the region, due to historical reasons, remained in that locality. In 2003, a group of farmers transitioned from conventional agriculture to organic farming following the creation of AGROPRATA (Rio da Prata Farmers' Association), an association of family farmers. Its members then began taking actions linked to environmental preservation [4].

Organic farming encompasses an agricultural system with sustainable techniques that do not allow the use of chemical inputs (fertilizers, pesticides) either for fertility enhancement or pest control [5]. The application of fertilizers can lead to increased doses of ionizing radiation in the population because fertilizers contain natural radionuclides from the uranium and thorium series, and studies show that the raw materials used in the production of phosphate fertilizers have a high concentration of uranium [6;7].

Radionuclides can be incorporated through ingestion, and within the human body, they become a source of internal exposure. Therefore, identifying and quantifying the radionuclides present in food is an important step in environmental monitoring and radiological protection. From this perspective, the activity concentrations of ^{40}K, ^{226}Ra, ^{228}Ra and ^{228}Th were calculated in six samples of organic fruits and vegetables grown in PEPB.

2. MATERIALS AND METHODS

Six foods - avocado, cabbage, carrot, persimmon, pineapple and red onion - grown at PEPB were purchased at an organic market. At the Laboratory of Environmental Analyses and Computational Simulation/Federal University of Rio de Janeiro (LAASC/UFRJ, its Portuguese acronym), the edible parts were selected and processed to make them homogeneous, without dehumidification, i.e., in natura. The samples were stored in a 500 mL polypropylene container sealed with plastic glue (to prevent ^{222}Rn leakage), the masses were measured on a Gehaka balance (model BG 4000) with an accuracy of ± 0.01 g, and the containers were wrapped in PVC film for storage in a freezer at a temperature of - 18ºC during 45 days to ensure secular equilibrium [8, 9]. In the event of incidents, it is recommended to analyze fresh food, since the moisture content can reach 95%, which implies efficiencies similar to those of water, with deviations of less than 1% for all energies [10]. Figure 2 shows two samples prepared for analysis.

Figure 2: Organic pineapple and persimmon samples

Source: Authors.

The gamma-ray spectra were obtained using a Canberra vertical coaxial HPGe detector, model GC3020, with a relative efficiency of 30% and an energy resolution of 1.816 keV for the 1332.5 keV peak of ^{60}Co. The multichannel system used was a Canberra DSA 1000 (Digital Spectrum Analyzer) with 8192 channels.

The counting time to obtain the area under the photopeak of interest of samples and background was 28,000s [11]. The background was determined by using an empty container identical to the one used to store the samples. To record the spectrum on the HPGe detector, the containers with samples, prepared as described above, are placed on a polystyrene support (absorber) of approximately 2 mm thickness. Figure 3 shows the data acquisition system.

Figure 3: Data acquisition system: (a) detector shielding and sample counting place; (b)dewar**;** (c) multichannel DAS 1000

Source: Authors.

The energy calibration of the detection system was carried out with sources with gamma emissions that coincide with or are close to the areas of interest in the spectrum. The certified standards used were ^{137}Cs, ^{60}Co, ^{22}Na and ^{40}K, totaling 5 experimental points of energy 661.6 keV, 1173 keV, 1332.5 keV, 1274 keV, and 1460 keV. The only certified source that matches the emission energy studied is ^{40}K, while the other energies are determined from the calibration curve adjustment, starting at channel zero and energy 0 keV and passing through all the energies of the calibration sources mentioned. Verification of the calibration and quality control was performed using a 500 ml beaker containing a sample in secular equilibrium from the Laboratory Intercomparison Program - Institute of Radiation Protection and Dosimetry. The emissions tracked in this study are from the nuclides ^{212}Pb, ^{214}Bi, ^{228}Ac and ^{40}K, with gamma emission at energies of 238.6 keV, 609 keV, 911 keV and 1460 keV, respectively. These radionuclides have been selected on the basis of their probability of emitting gamma radiation at these energies. Table I shows the radionuclides of interest, the transition isotopes (progeny), the gamma-ray energy lines used to determine the activity concentrations of ^{228}Th, ^{226}Ra, ^{228}Ra and ^{40}K, and their corresponding emission probabilities.

Table I: Transition isotopes and emission probability

Radionuclides	Transition isotopes	Energy (keV)	P_γ (%)
^{228}Th	^{212}Pb	238.6	43.6
^{226}Ra	^{214}Bi	609	45.5
^{228}Ra	^{228}Ac	911,1	25.8
^{40}K	-	1460	10.66

The activity concentrations of ^{226}Ra, ^{228}Ra and ^{228}Th were determined by the gamma energies emitted by the decay products ^{214}Bi, ^{228}Ac and ^{212}Pb, respectively, after secular equilibrium was established. The ^{40}K is a single gamma emitter at 1460 keV. The condition of secular equilibrium is when the half-life of the parent radionuclide is much longer than the half-life of its daughter radionuclides. The consequence is equal activity. [9, 11].

The ^{222}Rn, a gaseous descendant of ^{226}Ra, can easily escape the sample, which prevents the secular equilibrium activity of ^{226}Ra with ^{214}Pb and ^{214}Bi within the sample. For these radionuclides to reach equilibrium with ^{226}Ra, five to seven half-lives of ^{222}Rn (3.82 days each) are required, or approximately 30 days. Some scientific literature questions the 30 days, considering it too short to establish equilibrium. Thus, the 45 days have been adopted in more recent articles for analytical safety [9]. This time allows for the stabilization of the short-half-life decay products ^{228}Ac (6.15 hours) and ^{212}Pb (10.6 hours), which enables the detection of ^{228}Ra and ^{228}Th [11].

The LabSOCS computer programme was used to simulate efficiency values according to energy, as well as self-attenuation corrections. The simulation eliminates the requirement for radioactive sources when determining efficiency curves. The energy values were selected in advance, with intensities in keV of: 45, 60, 80, 100, 150, 200, 300, 500, 700, 1000, 1400 and 2000. The geometry was reproduced in LabSOCS software by inserting the sample characteristics and the sample holder (polypropylene container, 500 mL). As already mentioned, the moisture content in some vegetables implies efficiencies similar to those of water, indicating a reduced effect of self-attenuation, especially for the energy range used, 238.5 keV-1460 keV.

The efficiency simulation software calculates the densities. A geometry was generated for each of the samples due to the difference in density, which ranged from 0.90 to 1.04 g/cm^3 and an efficiency curve was obtained for each of them.[11].

The radionuclide activity concentration was calculated according to Equation 1 [12].

$$AC = \frac{N_L}{\varepsilon . P_\gamma . t . m} \quad (1)$$

Where AC is the specific activity concentration of the radionuclide (Bq.kg^{-1}); N_L is the net area under the photopeak of interest; ε is the detection efficiency of the system at the energy of interest; Pγ is the transition probability of the measured gamma ray (γ); m is the mass of the sample (kg) and t is the counting time (s).

Equation 2 was used to calculate the minimum detectable activity (MDA) per unit mass (Bq. kg^{-1}) [13].

$$MDA = \frac{k^2 \pm \sqrt{8k\sigma}}{\varepsilon . m . t . P_\gamma} \quad (2)$$

Where σ is the standard deviation of the background radiation from the shielding plus the sample holder measured over a time, t is the measurement time (s), m is the sample mass(kg), P_γ is the probability of emitting a certain energy; ε is the energy detection efficiency and k is the percentage of measurements outside a defined interval from the mean value [defined as 5%, k= 1.96].

The effective dose was calculated based on the annual food consumption published by the IBGE (2020) [14], according to Table II and the dose coefficient from the ICRP 119[15].

Table II: Annual food consumption

SAMPLE	ANNUAL CONSUMPTION (kg.year^{-1})
Avocado	3.212 (*)
Cabbage	0.438
Carrot	0.328
Persimmon	3.212 (*)
Pineapple	0.401
Red onion	2.482

(*) Foods not mentioned in the IBGE consumption table fall into groups according to their classification, i.e. other fruits.

Equation 3[16] was used to calculate the committed effective dose.

$$Def = Df \ x \ U \ x \ AC \tag{3}$$

Where Def corresponds to the annual effective dose (μSv.year^{-1}); Df is the dose coefficient published by ICRP 119 (Sv.Bq^{-1}); U is the amount of food consumed in a year (kg); AC is the specific activity concentration of the radionuclide in the food (Bq.kg^{-1})

3. RESULTS AND DISCUSSIONS

To calculate the activity concentration of the natural radionuclides ^{40}K, ^{226}Ra, ^{228}Ra and ^{228}Th and estimate the dose due to their ingestion, six organic vegetables grown in the PEPB by farmers affiliated with AGROPRATA, sold at certain fairs, were analyzed. The concentration of ^{40}K activity in all samples was above the minimum detectable level, allowing the effective dose to be estimated based on the consumption of the analyzed foods. Table III shows the obtained values for all samples analyzed.

Table III: Activity concentration of ^{40}K and annual effective dose

SAMPLE	ACTIVITY CONCENTRATION ($Bq.kg^{-1}$)	EFFECTIVE DOSE ($\mu Sv.year^{-1}$)
Avocado	74.45 ± 11.67	1.64
Cabbage	81.23 ± 12.78	0.25
Carrot	101.83 ± 15.47	0.23
Persimmon	33.10 ± 6.01	0.96
Pineapple	25.55 ± 5.18	0.08
Red onion	64.80 ± 10.42	0.56

The highest activity concentration of ^{40}K was obtained for the carrot (101.83 ± 15.47) $Bq.kg^{-1}$, and the highest effective dose value, 1.64 $\mu Sv.year^{-1}$, was for avocado. The lowest activity concentration of ^{40}K and effective dose values were obtained for the pineapple, (25.55 ± 5.18) $Bq.kg^{-1}$ and 0.08 $\mu Sv.year^{-1}$, respectively.

Table IV shows the comparison between the results obtained for the concentration of specific activity of ^{40}K of some fresh samples from the present work and those from the reference literature.

Table IV: Comparison of 40K activity concentrations (Bq.kg -1)

SAMPLE	PRESENT WORK	REFERENCE [11]	PERCENTAGE DIFFERENCE (%)
Pineapple	25.55 ± 5.18	49.00 ± 5.16	- 47.9
Carrot	101.83 ± 15.47	119 ± 7.98	- 14.4
Cabbage	81.23 ± 12.78	94.94 ± 6.85	- 14,4

When the activity concentrations in this study were compared with the reference, it was found that carrots and cabbage showed the same percentage difference: -14%. The largest difference was observed for pineapple at -47%. Negative values indicate lower ^{40}K

activity concentrations in organic products compared to conventional ones. Although the author of the reference work analyzed samples in nature by gamma spectrometry, with the same counting time, the discrepancies can be explained because the samples analyzed by [11] are from conventional crops. The variation between organic and conventional management methods reflects the complexity of the interaction between factors that influence the mobility and migration of natural radionuclides from the soil to the plant.

Concentrations of ^{40}K activity were found in all samples analyzed, which was expected since it is a naturally occurring element found in most foods and is an important component of biological structure [17].

Table V shows ^{40}K activity levels from the literature with the range of values obtained in this study.

Table V: ^{40}K activity concentrations ($Bq.kg^{-1}$) references

SAMPLE	^{40}K ACTIVITY CONCENTRATION ($Bq.kg^{-1}$)	REFERENCES
Avocado	251 ± 7	[18]
Cabbage	975 ± .76	[19]
Carrot	507.5 ± 27.76	[19]
	398.85 ± 6.89	[20]
Persimmon	-	NA
Pineapple	33.63 ± 0.38* 351 ± 3.98**	[21]
Red onion	68 ± 16	[22]
All samples	25.55 ± 5.18 - 101.83 ± 15.47	Present study

*Fresh sample **Dry sample NA-Not available.

References ^{40}K activity concentrations ranged from (33.63 ± 0.38* to 975 ± 76) $Bq.kg^{-1}$. While in the present study, the variation was (25.55 ± 5.18 to 101.83 ± 15.47) $Bq.kg^{-1}$.

The concentration of ^{226}Ra activity was below the minimum detectable level in all samples. Concentrations of ^{228}Ra activity were only detected in cabbage and carrots.

Concentrations of ^{228}Th were detected in all samples, although at low activity concentrations. Table VI shows the activity concentration of ^{228}Ra and ^{228}Th and the annual effective dose for the analyzed samples.

The different activity concentrations of radionuclides in different types of plants are the result of various factors, including the propensity to accumulate certain types of radionuclides and the possibility that the same radionuclide may accumulate in a particular compartment of the plant. In addition, the ability of plants to uptake long half-life radionuclides is mainly controlled by their degree of chemical accessibility and the fact that they remain within reach of their roots [17, 23, 24].

Natural radionuclides are absorbed with other nutrients and accumulate in different organs, including edible ones. This uptake is complex and depends on various factors such as the plant species, the conditions and concentrations of radionuclides in the soil and agricultural practices that can cause a mechanical redistribution of radionuclides. It is also important to note that plants have variable nutrient requirements throughout the year [25, 26].

Table VI: ^{228}Ra and ^{228}Th activity concentration and annual effective dose

SAMPLE	ACTIVITY CONCENTRATION (Bq.kg-1)		ANNUAL EFFECTIVE DOSE (μSv.year^{-1})	
	^{228}Ra	^{228}Th	^{228}Ra	^{228}Th
Avocado	-	0.28 ± 0.15	-	0.06
Cabbage	2.84 ± 1.19	0.94 ± 0.49	0.86	0.03
Carrot	1.31 ± 0.57	1.34 ± 0.68	0.30	0.03
Persimmon	-	1.04 ± 0.54	-	0.24
Pineapple	-	0.56 ± 0.29	-	0.02
Red onion	-	0.99 ± 0.51	-	0.09

Natural radionuclides are absorbed with other nutrients and accumulate in different organs, including edible ones. This uptake is complex and depends on various factors such as the plant species, the conditions and concentrations of radionuclides in the soil and agricultural practices that can cause a mechanical redistribution of radionuclides. It is also important to note that plants have variable nutrient requirements throughout the year [25, 26].

The root uptake of radionuclides depends on the state of the metal in the clay-soil particle-minerals mix and its availability to plants. In contrast, metals present in the atmosphere are deposited on various surfaces of the plant body. The deposition concentration varies over time due to wind, rain and the washed off metals reach the soil and become available for plant uptake [27].

Potassium is a macronutrient, the name given to the nutrient that plants absorb in significantly more quantity than micronutrients. It is an element that is actively involved in enzyme activation, stomatal activity, photosynthesis, sugar transport, starch synthesis, protein synthesis, as well as water and nutrient transport. Several factors influence potassium absorption such as: soil moisture, higher soil moisture generally means greater availability of potassium as it increases its movement to the plant roots and improves availability; aeration and oxygen levels in the soil, air is necessary for root respiration and potassium absorption; soil temperature, as it increases the physiological activity of the plant which leads to greater absorption of the element [28].

Despite the limitation of a small number of samples due to the difficulty of obtaining the products, as they come from family farms and are not always available for sale, the results contribute to the preliminary compilation of data on the activity concentrations of radionuclides present in these organic vegetables grown in the PEPB, which are frequently consumed by visitors to the site, and the doses to which they are exposed as a result of their consumption.

The activity concentrations of ^{226}Ra showed values below the minimum detectable activity. The levels of activity concentration of ^{228}Ra and ^{228}Th ranged from (1.31 ± 0.57 to 2.84 ± 1.19) $Bq.kg^{-1}$ and (0.28 ± 0.15 to 1.34 ± 0.68) $Bq.kg^{-1}$, respectively.

The extremely low activity concentrations of these radionuclides may suggest the absence of chemical fertilizers, since these can increase their concentrations.

4. CONCLUSIONS

Activity concentrations of ^{40}K were found in all samples, while for radionuclide ^{226}Ra, the values were below the minimum detectable level in all samples. Concentrations of ^{228}Ra and ^{228}Th were very low, resulting in a low committed effective dose. It is fundamental to note that the products marketed by AGROPRATA are certified, which means that they are inspected by specific entities. In this sense, the authors of this research are continuing their studies on the vegetables produced in the region, including soil analysis, to verify if the data obtained can suggest the absence of artificial fertilizers and also the concentration levels of natural radionuclide activity in the soils where organic food is grown. The estimated values for doses due to ingestion are within the limits set by the ICRP 119, which is 1 mSv/year, for individuals in the general public, upon which the ANSN standard NN 3.01 [29] is based. Thus, the consumption of these organic foods grown in the PEPB is radiologically safe.

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CONFLICT OF INTEREST

All authors declare that they have no conflicts of interest.

DATA AVAILABILITY STATEMENT

The authors declare that the data supporting the results of this study are available in the article. Derived data supporting the conclusions of this study are available upon request from the corresponding author.

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