BJRS

BRAZILIAN JOURNAL
OF
RADIATION SCIENCES
09-01A (2021) 01-16

Evaluation of radon, radium and gamma radiation at the environmental preservation area of Passaúna river, Brazil

Martin[a] A.C., Paschuk[a] S.A., Correa[a] J.N., Del Claro[b] F., Souza Filho[c] O.A.

[a] *Federal University of Technology – Paraná, 80230-901, Curitiba, PR, Brazil*

[b] *Positive University,81280-330, Curitiba, PR, Brazil*

[c] *CPRM -Geological Survey of Brazil, 80020-926, Curitiba, PR, Brazil*

alinemartin@alunos.utfpr.edu.br, spaschuk@gmail.com

ABSTRACT

The objective of this research was to evaluate the levels of natural radiation in the Environmental Protection Area (EPA) of the Passaúna River (Campo Largo region) in the state of Paraná, Brazil. Gamma radiation measurements were obtained with the gamma spectrometer, and for the measurements of radium and radon concentrations in soil and water there were collected 4 water samples from Passaúna's River, 2 well water samples from the region, 2 sample of a secondary lake and 4 soil samples, which were analyzed using the AlphaGUARD monitor and its accessories. The results show that the gamma spectrometric measurements found values ranging from 0.052 to 0.204 µSv / h. However, the results of the radon concentrations in the soil were significant, since the values obtained varied from 110 ± 1.30 kBq/m³ to 9 ± 0.80 kBq/m³, which results can be compared with the radon and radio measurements obtained in the wells water, which indicated values higher than the national limit of 0.5 Bq/L, established by the Brazilian Ministry of Health, and the World Health Organization. The values in the lake water were lower the limits. More measurements are needed to map the region to understand the radiation levels and to correlate water and soil measurements with the geological features of the place.

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

ISSN: 2319-0612
Accepted: 2020-11-15

1. INTRODUCTION

The radioactivity present in the Earth's crust has a predominantly natural origin, however, there is a portion coming from artificial sources. Natural radioactivity comes mainly from cosmic rays and emissions related to decays of natural radionuclides. Exposure to ionizing radiation from artificial sources is represented mainly by medical exposures, including nuclear medicine radionuclides, and consumer products [1].

Four natural isotopes of radium ^{223}Ra (half-life of 11.4 days), ^{224}Ra (half-life of 3.64 days), ^{228}Ra (half-life of 5.75 years) and ^{226}Ra (half-life of 1600 years) are produced in the natural series of uranium and thorium decay and could be found in natural soil at average activity, which vary from 67 to 17 Bq/kg [2]. Been more specific: ^{226}Ra, which can be found in rocks, soil, water and minerals, is considered as an important source of human exposure to ionizing radiation and its decay mainly by means of alpha emission originates ^{222}Rn isotope, which is also radioactive [3].

Among the natural radionuclides present and produced in the earth's crust radon requires special attention because it is a radioactive element that presents in the gaseous form, thus possessing a greater mobility and reaching more easily the atmosphere. Radon is a colorless, odorless and tasteless noble gas. When it is inhaled, because of its short half-live, the daughter isotopes of metals could be very easy adhere to the lung tissue and thus emitted radiation of their decay is deposited in a certain region / thickness of the lung tissue [4]. There is an important relationship between the human exposure to radon and lung cancer occurrence, which results in statement that radon is considered the second cause of such type of cancer [5].

The radon is also considered as the main source of human expose to radiation. Depending on the region it may contribute more than 50% of the average annual dose. Indoor standards are based on the concentration of radon in the air of dwelling and working places as well as there are international norms and regulatory recommendations concerning the limits for the radium concentration in water, soil and building materials since it is one from the principle alpha emitting natural radioisotopes [6].

Exposure to natural radiation is unavoidable, since it is present in any environment, but it is possible to weave circumstances in order to reduce human exposure. In this sense, there are norms and regulations established by specialized agencies that limit relative concentrations and / or doses. In Brazil there are no specific regulations regulating the limits of exposure to natural gamma radiation. Regarding the activity of radionuclides in water, the Brazilian Ministry of Health in 2017 published the Consolidation Ordinance No 5, which in article 38 of Chapter 5 of Annex XX determines that the total alpha activity values of a water sample should not exceed 0.5 Bq/L [7], which is congruent with recommendations of the World Health Organization [8].

The Environmental Protection Area of Passaúna is under special interest in relation to natural radioactivity, since it is a water supplying source for approximately 22% of Curitiba, Brazil. Even under restrictions in relation to the human occupation of the soil, the number of inhabitants in this area increased considerably in the last years, which justifying analysis regarding the concentration of natural radionuclides in the region.

The control of soil occupation in area of water springs is focused on water quality issues provided to the population. However, the control of soil occupation does not guarantee soil or water quality, due to factors related to the natural soil / water characteristics of the region. There are previous conditions of use that may have contaminated the environment or even some unknown contaminant and unregistered factors that may influence this issue.

The general objective of this research is to evaluate the natural radioactivity in the region of the Passaúna EPA well in Campo Largo / RMC, verify the concentrations of radon and radium in lake and well waters in the Passaúna spring region, as well as the radon concentration of the soil and to analyze the doses related to the natural gamma radiation in the region of the Passaúna.

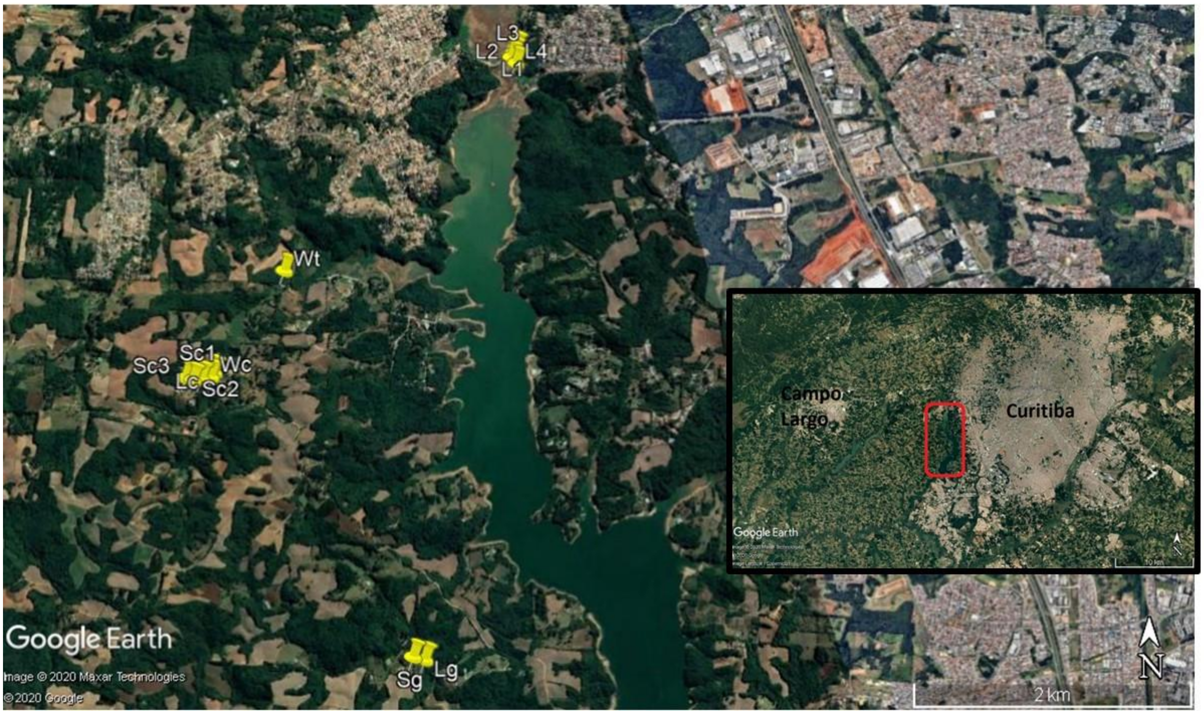
2. MATERIALS AND METHODS

The research was carried out in the Environmental Protection Area (EPA) of Passaúna, in the state of Paraná, in an area that includes the municipalities of Curitiba, Campo Largo, Araucária and Almirante Tamandaré. The main focus of the measurements was the EPA of the Passaúna located in the Campo Largo region. Samples of water (main and secondary lakes) and well water were collected from EPA region. Soil measurements were taken at sites correlated to the water samples under study. Gamma spectrometry measurements were also performed in the region.

Radon and radium measurements were performed in the second semester of 2018. Data collection for gamma radiation was extended to the first semester of 2019. After the samples were collected, they were analyzed in the Laboratory of Applied Nuclear Physics (LFNA) of the Federal University of Technology – Paraná, UTFPR.

Gamma measurements were preliminarily performed at the Passaúna. The most significant values of the gamma spectra measurements were considered as indicators of the points of interest for radium and radon monitoring. The area that contains these points of interest is composed of small farms. The specific points measured in this area were chosen considering the ease of access and support of the local population. Fig. 1 shows a map with the points where the measurements were made.

Figure 1: *Map of Passaúna reservoir region (https://earth.google.com/web/@-25.4936045,-49.37738242,900.24344397a,14614.78843388d,34.999998y,0h,0t,0r) with the measurement point pins.*



Source: Adapted from Google Earth (2020)

2.1. Environmental Gamma Spectrometry

The gamma spectrometric measurements of this research were performed using a gamma spectrometer model AT6101C (Atomtex), which can be seen in Fig. 2. This spectrometer uses a NaI (Tl) scintillator with dimension of 63x63 mm associated with a photomultiplier tube.

Gamma spectrometric measurements were performed so that it was possible to cover a larger area. The equipment was positioned inside a van automobile at the same height as the user's chest. During the measurement the data collection was performed every 30s.

Figure 2: *Spectrometry equipment. A) Case for transportation of equipment; B) Power and electronic modules; C) Gamma and neutron detectors*.

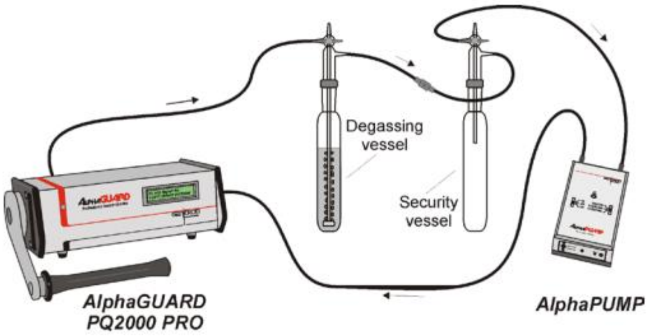
Source: Author

2.2. Environmental Radon and Radium Measurements

Measurements that identified radium and radon presence were performed using the AlphaGUARD monitor equipment (Saphymo, current Bertin Technologies SAS). AlphaGUARD uses ionization chamber to detect alpha particles emitted from its gaseous content. The apparatus also has the diffusion mode of operation, where the gas diffuses through a glass fiber filter into the ionization chamber. AlphaGUARD ionization chamber measures concentration in the range from 2 to 2,000,000 Bq/ m³. It has to be noted that AlphaGUARD detector is not able to discriminate the radioactivity of radon isotopes separately, which means that measured values represent the total alpha radioactivity activity of radon isotopes in the sample.

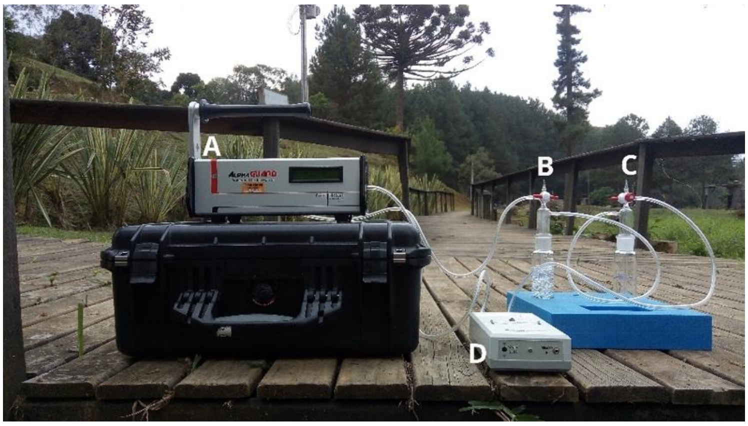
The measurements that identified the presence of radium and radon in water were performed using the AlphaGUARD equipment with the specific kit glass vessels for water measurements. The AquaKIT consists of an aeration vessel, safety vessel and connecting hoses as it could be seen in Fig. 3 and 4, which show the circuit used in radon measurements with AquaKIT. The photograph presented in Fig. 4 illustrates the measurement relative to one of the lakes monitored in this research.

Figure 3: *Scheme of the AquaKIT arrangement.*



Source: AlphaGUARD manual

Figure 4: *Schematic of the AquaKIT arrangement used for water measurements. A) AlphaGUARD detector; B) water sample; C) safety glass; D) Alpha Pump.*



Source: Author

The results concerning the radon activity the air that passes through the measuring system with the water sample subsequently were recalculated to the values for radon concentration in the water according to protocol based on the equipment manual.

Used protocol for these measurements of radon in water was 1min flow, which means that the coupled software integrates the data in periods of 1 minute. The flow of the pump providing air circulation by the sample and equipment was of 1L/min, according to mentioned protocol. Each measurement had total time of about 1 hour. The initial 10 minutes of the measurements were performed with the pump switched off to measure the background intrinsic to the system. Then the pump was tern on for 40 minutes when the radon isotopes ^{222}Rn and ^{220}Rn were released from the water sample into circulated air.

2.1.1. Well Water Measurements

The water samples from two semi artesian wells were collected in to plastic bottles, avoiding that the sample was agitated at the moment of extraction. Excess air was withdrawn from the vial which was then sealed until the measurement. The bottles were held tightly closed during the interval between measurements, and after each measurement the excess air was withdrawn again and the sample was resealed. The interval between measurements is described in Table 1.

Table 1: Measurements chronogram dates.

Measurement 1	Just after water sample collection (time zero)
Measurement 2	After 5th day
Measurement 3	After 10th day
Measurement 4	After 16th day
Measurement 5	After more than 30 days from water sample collection (secular equilibrium)

The samples were measured with an interval of 3 to 6 days in order to adjust a curve representative of the radon concentration activity over time. The final measurement is performed after approximately 30 days when the radon activity is the same as its parent radium isotope, indicating the secular equilibrium.

2.1.2. Lake water measurements

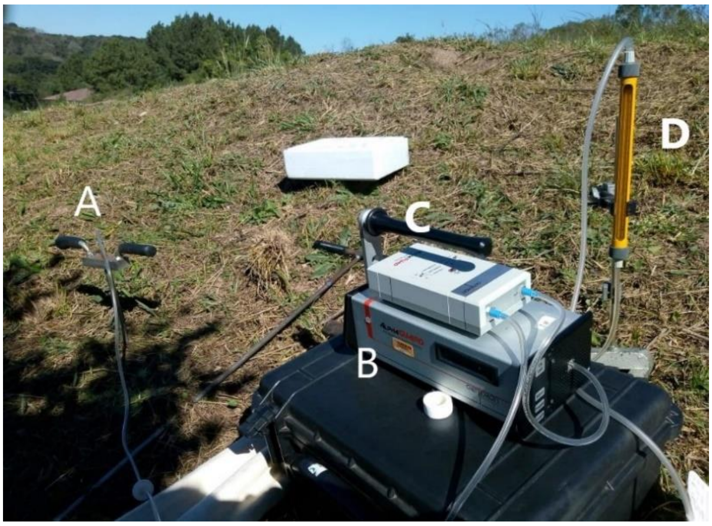
Samples were collected from three different lakes in the region: Lake (reservoir) Passaúna with 4 sampling points and 2 small secondary lakes, with one sampling point each. The samples of these lake water were collected following the well water protocol as described previously.

For the lake samples, only indirect radium measurements were chosen, taking into account that radon measurements alone would not be relevant, due the size of the lakes and the agitation of the water. Therefore, for lake waters radium analyzes were performed after approximately 30 days, when the secular equilibrium was reached.

2.3. Soil measurements

Soil measurements were also performed using the AlphaGUARD detector, now associated with a soil survey probe inserted into the soil reaching the depth of approximately 1 meter. Before the actual data acquisition, background measurements were performed during 10 min. After background evaluation, the effective measurements were taken. Soil research was conducted in four different areas in the smaller lakes' region. The arrangement used for the soil measurements can be observed in Fig. 5.

Figure 5: *Arrangement for radon in soil measurements. A) probe inserted at the depth of 1m; B) AlphaGUARD radon detector; C) Alpha Pump; D) Rotameter.*



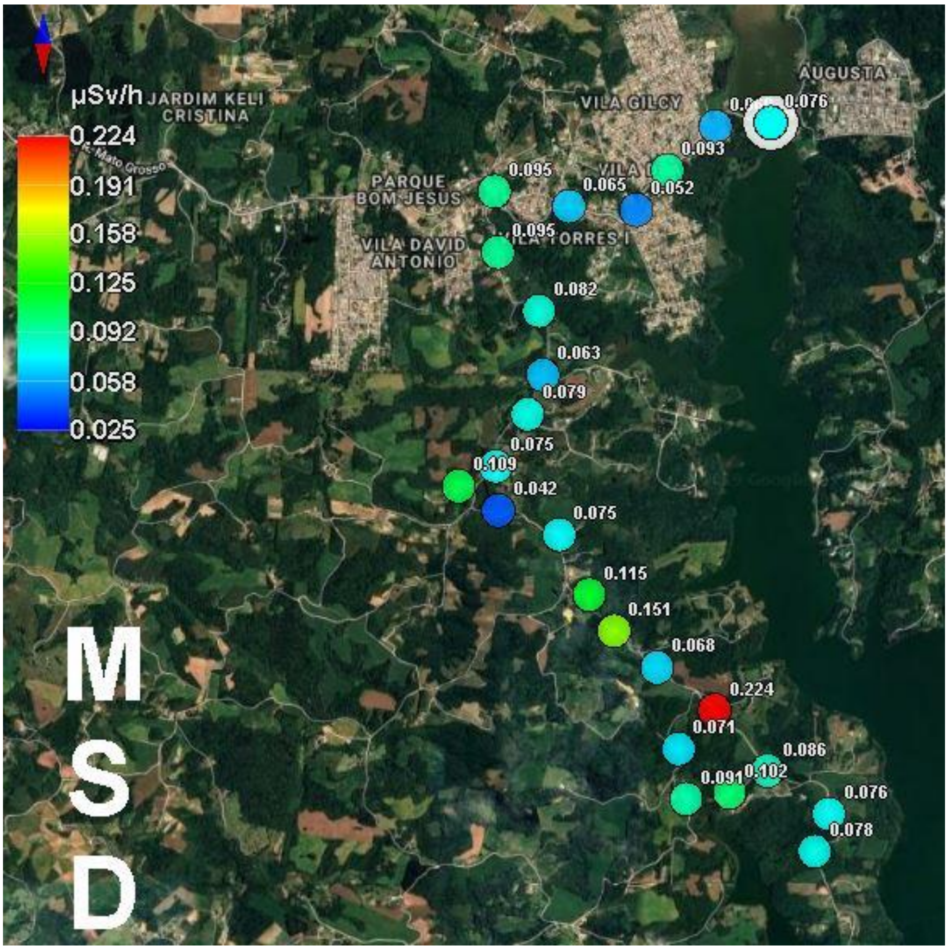
Source: Author

3. RESULTS AND DISCUSSION

3.1. Environmental Gamma Spectrometry

The results of the environmental gamma spectrometry measurements of the Passaúna region located in the municipality of Campo Largo found dose rate values of 0.052 to 0.204 µSv / h. In Figure 6 it is possible to observe the values provided by the spectrometer, in a sequence of points (local) in the aero photogrammetric map.

Figure 6: *Gamma spectrometry values located on the map of the Passaúna region.*



Source: Author

Although there is no standard that determines the limit of exposure to natural gamma radiation these values are important to observe a general overview of radiation levels in the region.

3.2. Well Water Measurements

Water measurements were performed from two different wells, named Wt and Wc, already mentioned in the previous item. The values of radon activity can be seen in Table 2.

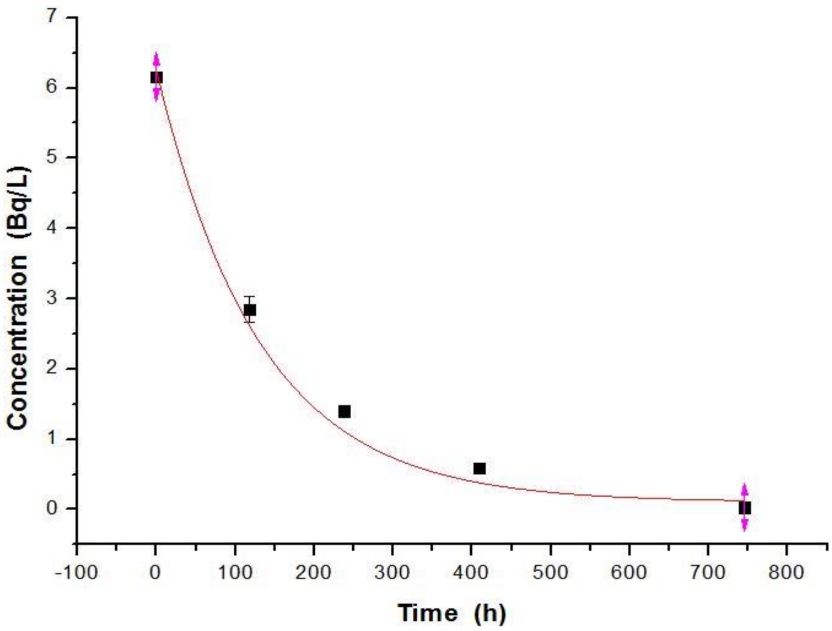
Table 2: Results of radon measurements in well water.

	Wt		Wc	
Order of	**Measurement**	**Error**	**Measurement**	**Error**

Measurements	**(Bq/L)**		**(Bq/L)**	
1	6.2	0.063	2.09	0.04
2	2.8	0.18	1.2	0.047
3	1.4	0.062	0.96	0.038
4	0.59	0.032	0.38	0.025
5	0.031	0.019	0.23	0.016

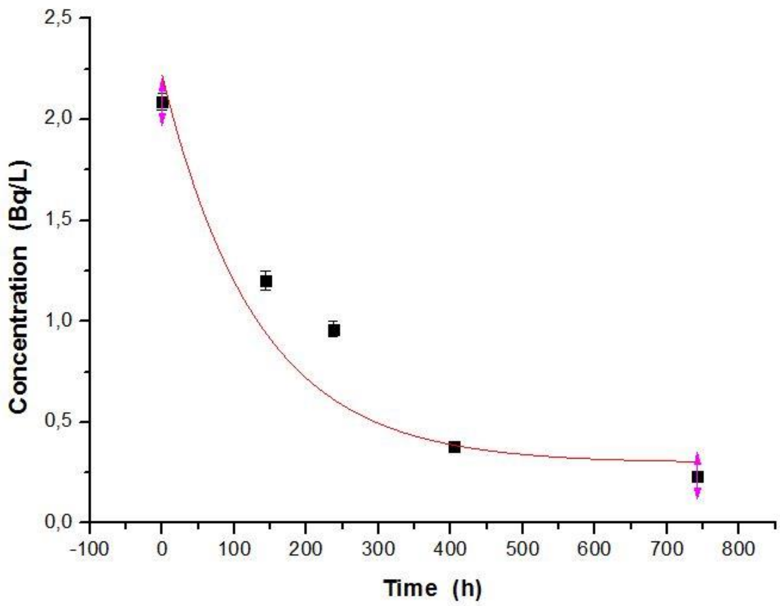
Due to the fact that the measurements were carried out in 5 different stages, it is possible to present a curve of radon decay in the water sample from its collection until reaching the secular equilibrium. These curves are shown in Figs. 7 and 8.

Figure 7: *Decay curve in Bq / L referring to sample Wt.*



Source: Author

Figure 8*: Decay curve in Bq / L referring to sample of Wc.*



Source: Author

The results showed very high values of radon activity at the time of sample collection. For well Wt the result was of 6.2 Bq/L, which is more than 10 times above the established limit of the Brazilian Ministry of Health [7] and the World Health Organization [8]. But approximately 16 days later, the levels of radon activity in water samples were already below or very close to the limit value. This indicates the presence of diluted radon and means that these sources of water are not safe for immediate consumption but can be consumed after reaching secular equilibrium.

3.3. Lake water measurements

Measurements were performed using water samples from the 4 sample collection points of the Passaúna dam storage (Lp). The results of the measurements of the samples relative to the radium, made indirectly in the secular equilibrium between radon and radium, can be observed in Table 3.

Table 3: Results of radon/radium measurements of water samples collected at the Passaúna reservoir dam Lp.

	Measurement (Bq/L)	Error
Sample Lp1	0.180	0.015
Sample Lp2	0.260	0.027
Sample Lp3	0.240	0.023
Sample Lp 4	0.200	0.025
Lp Average	0.220	0.046

Found values are within the limits established by the WHO-Guidelines for drinking-water quality [8].

The results of the radium measurements in the secondary lakes (Lc and Lg) are shown in Table 4. The measurements in the Lg region were purposely performed near the area where gamma spectroscopy values were found elevated.

Table 4: Results of radium activity measurements in water of secondary small lakes of the region.

	Measurement (Bq/L)	Error
Lc	0.18	0.047
Lg	0.39	0.031

Both measured radium activity concentration values are below the established limits. It could be noticed that although the gamma radiation values of the Lg lake region are high, the measurement of the radium concentration in the water samples is below the global alpha limit.

3.4. Soil measurements

The soil radon measurements Sc1, Sc2 and Sc3 were performed in the region near the well analyzed WC. The Sg soil measurement was performed near the region where gamma spectroscopy values were elevated. The values found in the soil measurements can be observed in Table 5.

Table 5: Results of soil measurements.

	Measurement (Bq/m³)	Error
Sc1	38.000	1400
Sc2	34.000	3200
Sc3	110.000	1300
Sg	9.200	830

It can be noticed that, although the results observed in the measurement of gamma spectrometry are higher comparing with other points in the region in general, the value of the radon concentration in the soil was lower than the other measured points.

4. CONCLUSIONS

It was possible to verify that the values of radon activity concentration in the well water samples were above the established limit, but after reaching the secular equilibrium, both analyzed samples had radon activity value within the limit. Both samples taken from the Passaúna dam and the samples from the secondary lakes presented the values of radium activity concentration within the limits. The gamma spectroscopy measurements of the region provided an average dose picture, the results showed no clear relationship between the highest dose rate values and the highest values of radium and radon concentration, but further measurements would be required to confirm or discard this correlation.

ACKNOWLEDGMENTS

The authors are very thankful to all colleagues from UTFPR who helped us to develop present research. Also, we would like to express our gratitude to CNPq, CAPES, CNEN and Fundação Araucária (Paraná St.) for financial support of this research project.

REFERENCES

[1] SCHAUER, D. A.; LINTON, O. W. NCRP report No. 160, Ionizing radiation exposure of the population of the United States, medical exposure—are we doing less with more, and is there a role for health physicists? **Health physics**, v. 97, n. 1, p. 1-5, 2009.

[2] INTERNATIONAL ATOMIC ENERGY AGENCY, The Environmental Behavior of **Radium: Revised Edition**, Technical Reports Series No. 476, IAEA, Vienna (2014).

[3] EISENBUD M.; GESELL, T. **Environmental Radioactivity from Natural, Industrial and Military Sources**. 4ª ed. California: Academic Press, 1997.

[4] TURNER, J. E. **Atoms, Radiation, and Radiation Protection**. 3ª ed. Wiley-VCH Verlag, 2007.

[5] WHO – World Health Organization. **Handbook on Indoor Radon**. A Public Health Perspective, Switzerland: WHO press,2009.

[6] UNSCEAR - **United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and Effects of Ionizing Radiation**, Anexo B, 2008. UNSCEAR Report to the United Nations General Assembly.

[7] BRASIL, Ministry of Health. Portaria de Consolidação nº 5, de 28 de Setembro de 2017. **Consolidação das normas sobre as ações e os serviços de saúde do Sistema Único de Saúde** (in portuguese)**.** Brasília, DF, set. 2017.

[8] WORLD HEALTH ORGANIZATION. **Guidelines for drinking-water quality**. World Health Organization, 2004.