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DETERMINATION OF THE ^{226}Ra, ^{228}Ra AND ^{210}Pb CONCENTRATIONS IN THE MINERAL WATER SPRINGS FROM WATER PARKS OF CAMBUQUIRA AND MARIMBEIRO, MINAS GERAIS AND ASSESSMENT OF THE COMMITTED EFFECTIVE DOSES.

Laíssa A. B. dos Santos[1] and Sandra R. Damatto[1]

[1]Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
lbonifacio@ipen.br, damatto@ipen.br

ABSTRACT

Due to the importance of human life, water quality must be controlled and a very important parameter are the limits of the natural radioactivity of the water consumed.In relation to therapeutic practices based on water intake the radionuclides ^{210}Pb , ^{226}Ra and ^{228}Ra when present, are of great importance because they contribute to the internal irradiation of individuals.The study areas of the present work were the Parque das águas of Cambuquira and Marimbeiro, located in Minas Gerais. Hence, the objective of this work was to determine the ^{226}Ra, ^{228}Ra and ^{210}Pb concentrations in the springs of these parks and evaluate the committed effective doses due to its consumption.The radionuclides were determined by a radiochemical procedure in four collections at different seasons of the year. The Concentrations ranged from 4 ± 1 mBq / L to 509 ± 32 mBq / L for ^{226}Ra, from 3.7 ± 0.1 mBq / L to 631 ± 27 mBq / L for ^{228}Ra and 5 ± 1 mBq / L to 60 ± 5 mBq / L for ^{210}Pb. The dose for adults from $1.21x10^{-01}$ mSv/y for ^{226}Ra, $3.18x10^{-01}$ mSv/y for ^{228}Ra and for ^{210}Pb $3.02x10^{-02}$ mSv/y.

1. INTRODUCTION

The human being is constantly exposed to some source of radiation, which can be of anthropogenic origin or of natural origin It is known that approximately 98% of the dose normally received by the population comes from natural radiation, there fore many studies are developed to assess the possible biological effects on human health when the individual is exposed to prolonged low doses of ionizing radiation [1,2]

However, several factors can modify the amount of dose received by each individual and among these is the consumption of drinking water.

According to the World Health Organization (WHO) the recommended effective dose comprising 1 year of consumption and use of drinking water is 0.1mSv [3].

Mineral water, in the Braziliam law No. 7.841 of 08.08.1945 is defined as: "Mineral waters are those from natural or artificially collected sources that have chemical or physical composition or physico-chemical properties distinct from common waters with characteristics that give them a medication action" [4].

In addition to chemical composition, these waters may be classified by other physical chemical characteristics, such as temperature and gases; they are also studied in the field of medicine.

Historical studies show that mineral waters were used since babylonian times as a form of therapy, but it was in Gália that it became known as the city that started the trade of mineral waters (medicinal), mainly cold waters. Although the indigenous people always use water for therapeutic purposes only in the nineteenth century that the use of these started in Brazil, in 1818, with the creation of the first Brazilian spa that should be governed by the statutes of Hospital of Caldas da Rainha, located in Portugal, since then the use of thermal waters has intensified [5].

Several studies have shown that periodic table the elements of that have isotopes can be found in water and a important characteristic of mineral waters are the high concentrations of ^{226}Ra and ^{222}Rn originally dissolved [6,7]. A study carried out in the city of Recife-PE determined values of ^{222}Rn in mineral water, varying from 5 ± 0.2 Bq / L to 373.2 ± 7.2 Bq /L [8]. Another study conducted in the state of São Paulo and Minas Gerais found an activity concentration of ^{222}Rn in the range of 0.02 Bq / L to 112.5 Bq / L [9]. For the ^{226}Ra concentrations in mineral waters determined in several areas of Brazil in a study performed by Godoy et al.[10] the values ranged from 0.002 Bq / L to 0.22 Bq / L.

It should be emphasized, however, that in relation to therapeutic practices based on water intake, immersion thermal baths and the use of medicinal sludges, the radionuclides ^{210}Pb and ^{228}Ra when present are of relevant importance, since they contribute in large part to the internal and external irradiation of the individuals.

The chemical element radio is an alkaline-earth metal, with atomic number 88, and which has no stable isotopes. Although approximately 25 radio isotopes have been identified, the natural isotopes of greater interest and in greater abundance are: ^{226}Ra coming from the natural series of ^{238}U, ^{223}Ra coming from the natural series of ^{235}U ,^{224}Ra and ^{228}Ra coming from the natural series of ^{232}Th. Radio behaves very similar to barium, not only because they belong to the same family in the periodic table, but mainly because of the similarity of their ionic rays, hence barium used as its chemical analogue. On the incorporation of these radionuclides, ^{226}Ra and ^{228}Ra present a behavior similar to calcium, a large part of the ingested radio is eliminated by fecal means, but the remaining tends to deposit in the bones [11,12].

Lead is a chemical element that has atomic number 82 and atomic weight 207. This element is rarely found in the elemental form, being considered a rare element because of its little abundance in the earth. There are three isotopes of lead occurring in the natural radioactive series ^{211}Pb, ^{212}Pb and ^{214}Pb, but only the ^{210}Pb, has a relatively long half-life compared to its other isotopes. Most of the ^{210}Pb, when inside the human body, has the affinity of to deposite in the skeleton [13].

The objectives of the present work were to determine the activity concentrations (mBq /L) of the natural radionuclides. ^{226}Ra, ^{228}Ra and ^{210}Pb in the mineral waters of Parque das águas Cambuquira and the Marimbeiro, both located in the state of Minas Gerais,for a period of 12 months with quarterly samplings and to evaluate the committed effective dose due to the ingestion of these waters containing these radionuclides.

2. MATERIALS AND METHODS

2.1 Study area

The city of Cambuquira is located in the south of the state of Minas Gerais state at the coordinates 21 ° 51 '30 "S and 45 ° 17' 28" W. The population of the city is estimated in 13,040 people in a territorial area of 246,380 km² [14]. The morphology of the city is defined by hills of soft slopes, where the predominant rock is a micaceous quartzite, weathered [16]. The town was once a farm named Boa Vista that belonged to three single sisters; when the sisters died the property passed into the hands of slaves. Fearing to lose the property, since

the local drew attention because the mineral water, the City Council of Campanha intervened considering the area of public utility area and the place then changed the name to Cambuquira. In 1890, taps were isolating the springs by Dr. Americo Werneck and chemist Frances Ch.Berthaud [15].

The Parque das águas of Cambuquira (FIG.1) concentrates the springs: Regina Werneck (Gasosa), which has three water , that in the present work were numbered as I, II and III, Souza Lima (Sulforosa), Fernandes Pinheiro (Férrea), Augusto Ferreira (Magnesiana) and Roxo Rodrigues (Litinada), the latter being used in a mineral water bottling unit in the city of Cambuquira. The park also has a lake, and a spa.

Figure. 1- Parque das águas of Cambuquira

The other study is the Parque das Águas of Marimbeiro (FIG.2) located 3 km from the center of the city of Cambuquira; the park presents three water taps that were denominated Marimbeiro 1, 2 and 3. Both parks are managed by CODEMIG [16]. A sample of a mineral water spring was also collected from a private property open to the population, near to the park of Cambuquira and called the source of Laranjal or Dico spring (FIG.3).

Figure 2. Parque das águas of Marimbeiro.

Figure 3. Laranjal or Dico spring

2.2 Sampling

Four representative collections of each season of the year (winter ,spring, summer and) were performed, from June 2016 to February 2017. The samples were collected in polyethylene bottles and acidified with 50% nitric acid until pH ≤ 2.0 in order to avoid adsorption of the radionuclides in the bottle walls [17].

2.3 ^{226}Ra, Ra-228 and ^{210}Pb determination

The radionuclides ^{226}Ra, ^{228}Ra and ^{210}Pb were determined using a sequential radiochemical procedure described in Oliveira [18] and Moreira [13]. The water samples were preconcentrated from 2 L to 1 L by evaporation in duplicate, and the radionuclides were initially precipitated with H_2SO_4 3 mol/L in basic pH of NH_4OH. The precipitate was dissolved with tri-acetic nitrile acid at basic pH and reprecipitated with acetic acid and ammonium sulfate as sulfate. The radionuclides ^{226}Ra and ^{228}Ra remained in the precipitate and were reprecipitated at basic pH as $Ba(Ra)SO_4$. ^{210}Pb, which remained in the supernatant was precipitated with Na_2CrO_4 as $^{210}PbCrO_4$. Both precipitates were filtered using glass fiber filter paper and the chemical recovery was determined gravimetrically. Activity Concentrations of the radionuclides ^{226}Ra and ^{228}Ra were determined by measurement of the gross alpha and beta, respectively, and ^{210}Pb by gross beta measurement through its ^{210}Bi decay product in a in a low back ground gas flow proportional detector.

2.4 Committed effective dose

the committed effective dose is the dose to the whole body incorporated in a given period and can be determined using the expression (1) :

$$\mathbf{H_e = C_n \times I \times FCD_e} \qquad \mathbf{(1)}$$

Where H_e corresponds to committed effective dose (mSv y^{-1}), C_n is the activity concentration of the radionuclide in water (Bq L^{-1}); I is the rate of water consumption in L per year (730 L y^{-1}), and FCD_e corresponds to the committed effective dose conversion factor due to the intake of the interest radionuclide wich were taken from the International Commission on Radiological Protection ICRP 119 [19].

3. RESULTS AND DISCUSSION

All analyzes were performed in duplicate, therefore the final results of the concentrations were expressed as arithmetic means and standard deviation.

The Souza Lima (Sulfurosa) spring was not analyzed in this work, because during the collection period it was under maintenance. In the autumn sampling of 2016 the spring Laranjal ou Dico was not sampled.

3.1 ^{226}Ra, Ra-228 and ^{210}Pb

A sample of drinking water from the city was collected at all stations in order to verify if the waters of the parks had characteristics different from that provided in public supply

FIG. 4 shows the activity concentrations of the radionuclides ^{226}Ra, ^{228}Ra and ^{210}Pb in mBq/L, in the four samplings performed and in all springin the Parque das águas of Cambuquira and Marimbeiro.

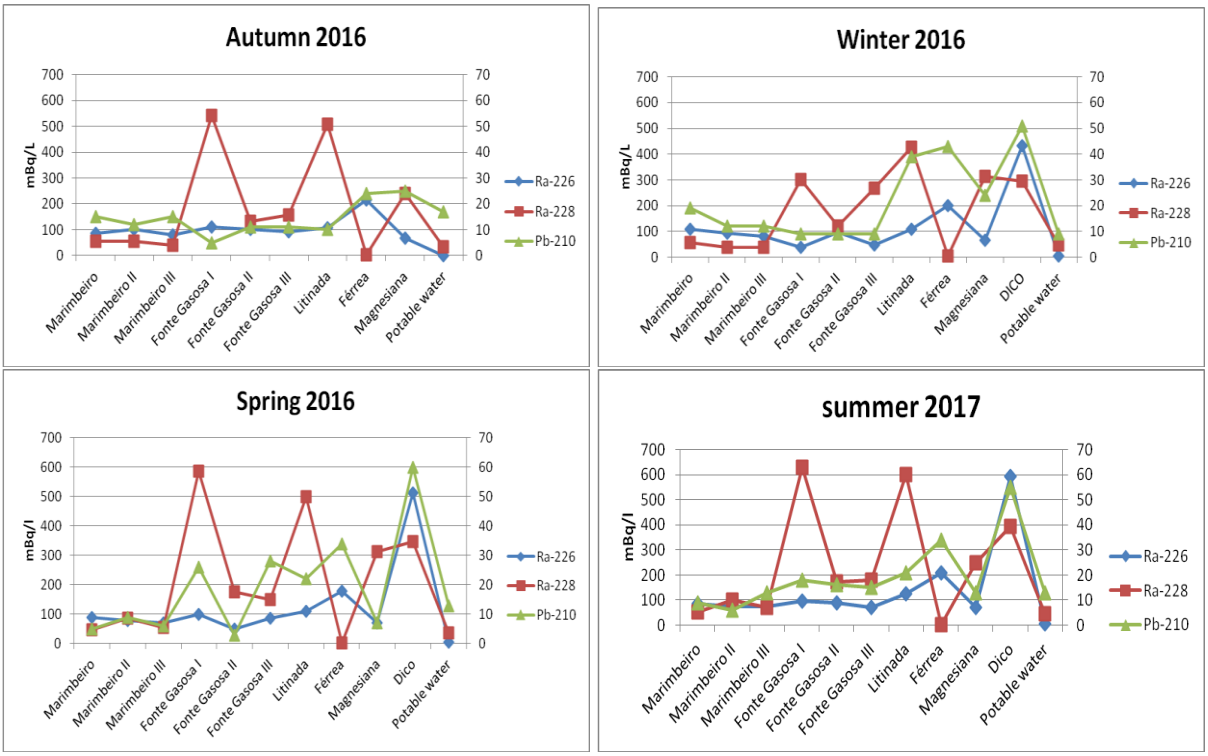

Figure 4 . Concentrations of ^{226}Ra, ^{228}Ra and ^{210}Pb radionuclides in mBq / L

The radionuclides ^{226}Ra and ^{228}Ra had the highest concentrations in the majority of the samples in relation to ^{210}Pb, because the of Ra isotopes, as well as those of U and Rn are those that present greater solubility in aquatic environments [20].

The spring that presented the highest concentration of ^{226}Ra, 509 ± 32 mBq / L, was the Laranjal or Dico in the sampling referring to the summer. For ^{228}Ra the highest concentration was also obtained in the summer collection, 631 ± 27 mBq / L, for the spring Gasosa I. For the radionuclide ^{210}Pb the spring that presented the highest concentration in all the collections was also the Laranjal or Dico with concentration of 60 ± 5 mBq / L referring to the spring

sampling. The collected drinking water sample showed values ranging from 4 ± 1 mBq/L to 6 ±1 mBq/L for ^{226}Ra, 37± 3 mBq/L to 47±7 mBq/L for ^{228}Ra and 9 ± 1 mBq/L to 17± 2 mBq/L for ^{210}Pb.
The values found in the majority of the spring for ^{226}Ra and ^{228}Ra are higher than those found in a study conducted with potable water from Italy, where the activity concentrations ranged from 0.50 mBq / L to 60.8 mBq / L for ^{226}Ra, with a maximum concentration for ^{228}Ra of 25.7 mBq / L [21]. However, when comparing the values obtained in the present study with the study conducted by NEGRÃO [22] where the mineral waters of Caxambu, MG, were studied,the springs Venâncio, Beleza, D. Isabel and Ernestina presented higher values.
For ^{210}Pb it was possible to observe that the values found are relatively higher than the values determined in the study that analyzed the waters of the Guarani Aquifer [23]. It was also observed that the concentrations of ^{210}Pb of the present study are within the range determinatted in mineral waters from Minas Gerais, that varied from 0.05 mBq /L to 110.6 mBq / L [24].

3.2 Committed effective dose

The values of effective doses (He) mSv / year obtained in this work are presented in FIGs, 5, 6, 7 and 8 for individuals of the public of 5 years, 15 years and adult for all the samplings and the figures also shows the dose limit of 0.1 mSv / year established by WHO.The highest committed effective doses obtained for the adult population were $1.21x10^{-01}$ mSv /y for ^{226}Ra referring to the spring Laranjal or Dico, in the summer, $3.18x10^{-01}$ mSv /y for ^{228}Ra referring to the spring Gasosa I also in the summer and for ^{210}Pb $3.02x10^{-02}$ mSv /y referring to the spring Laranjal or Dico in the spring of 2016. These values are higher than those found in the mineral waters of the Municipal Spa of João de Águia Pupo (SP), but when compared to the effective doses determined in the Caxambu park, in their totality the results are smaller [22].

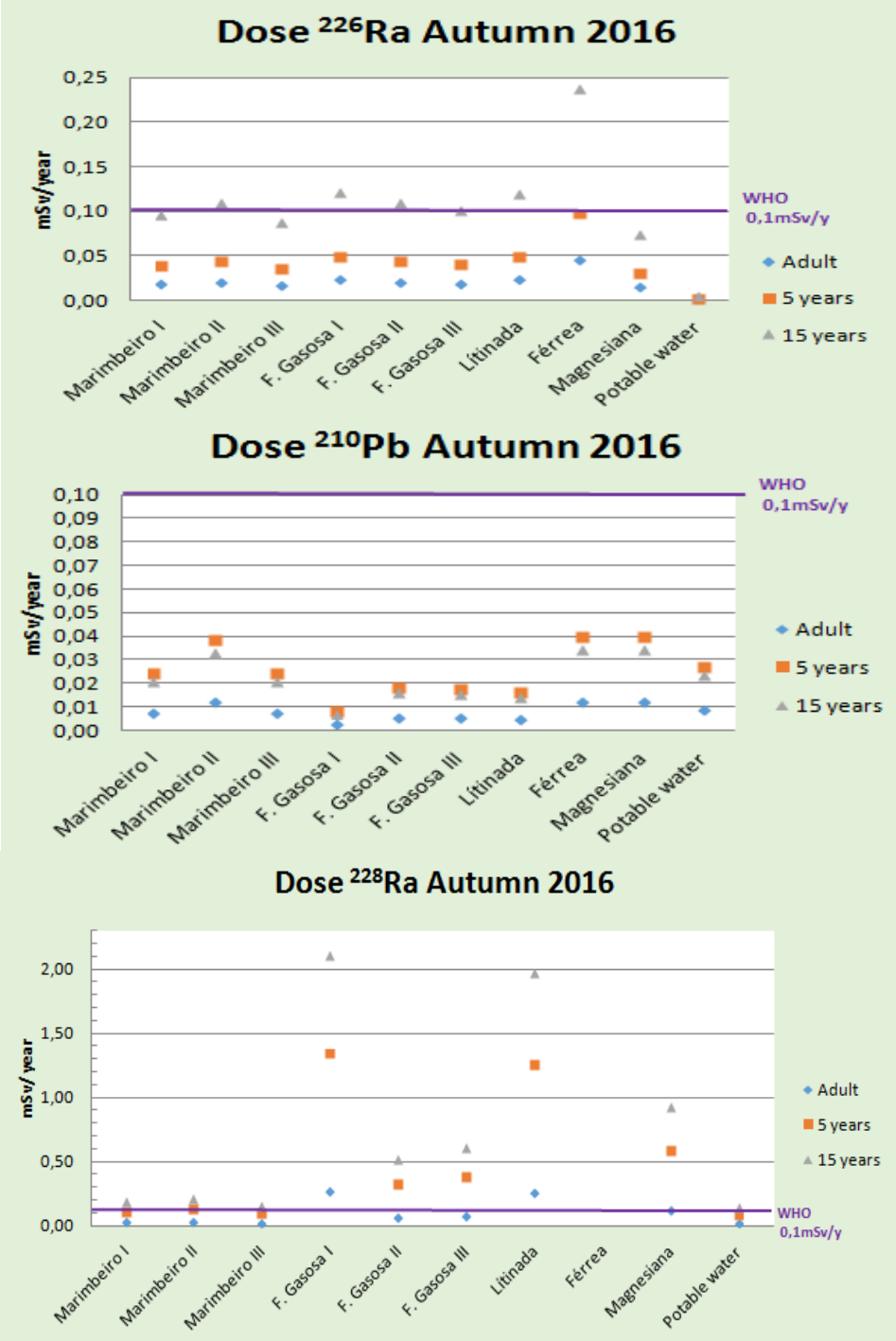

Figure 5- Committed Effective dose for ^{226}Ra, ^{228}Ra and ^{210}Pb in the mineral waters of the Parque das Águas of Cambuquira and Marimbeiro in the autumn of 2016

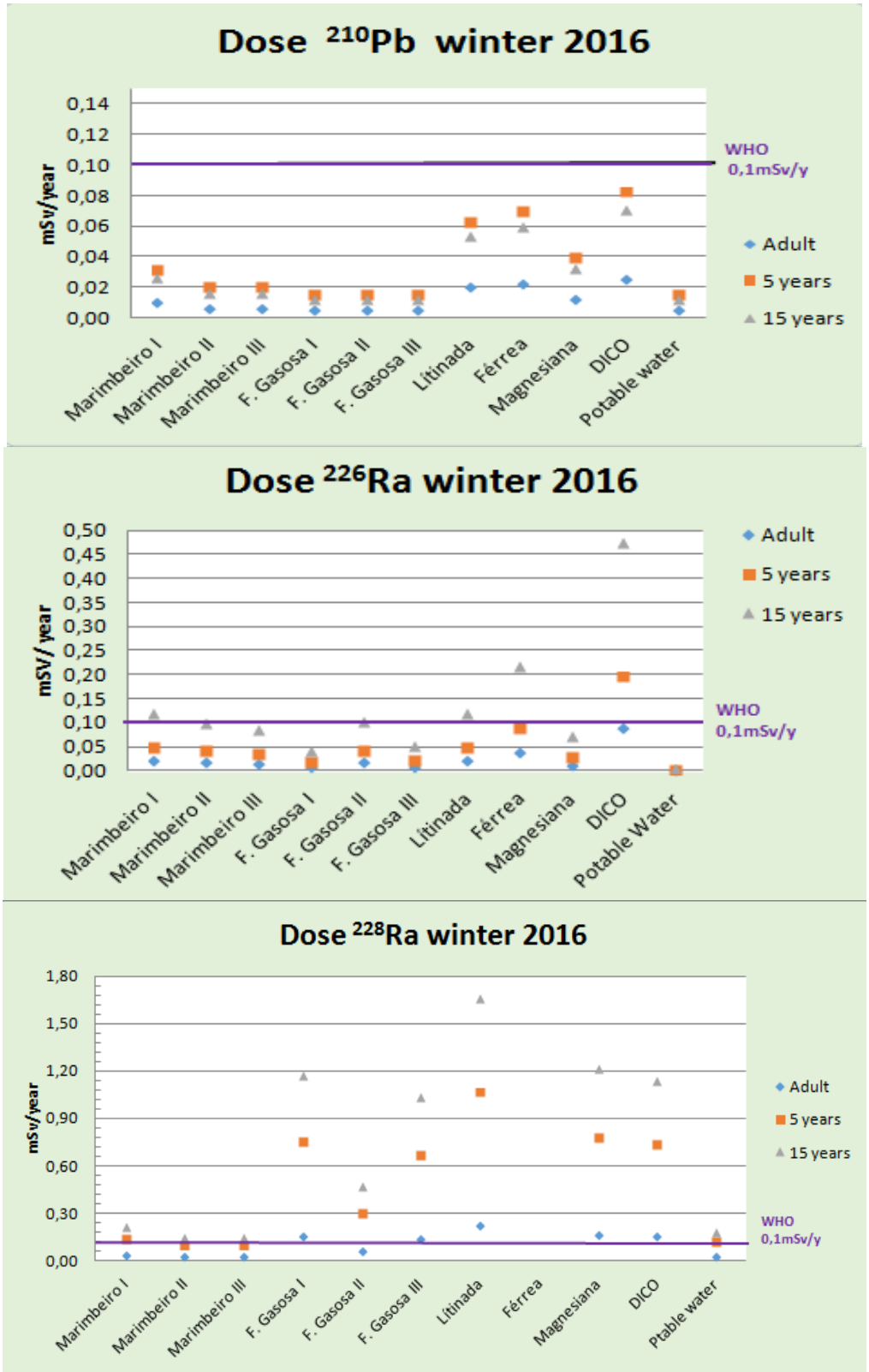

Figura 6 - Committed Effective dose for ^{226}Ra, ^{228}Ra and ^{210}Pb in the mineral waters of the Parque das Águas of Cambuquira and Marimbeiro In the winter of 2016

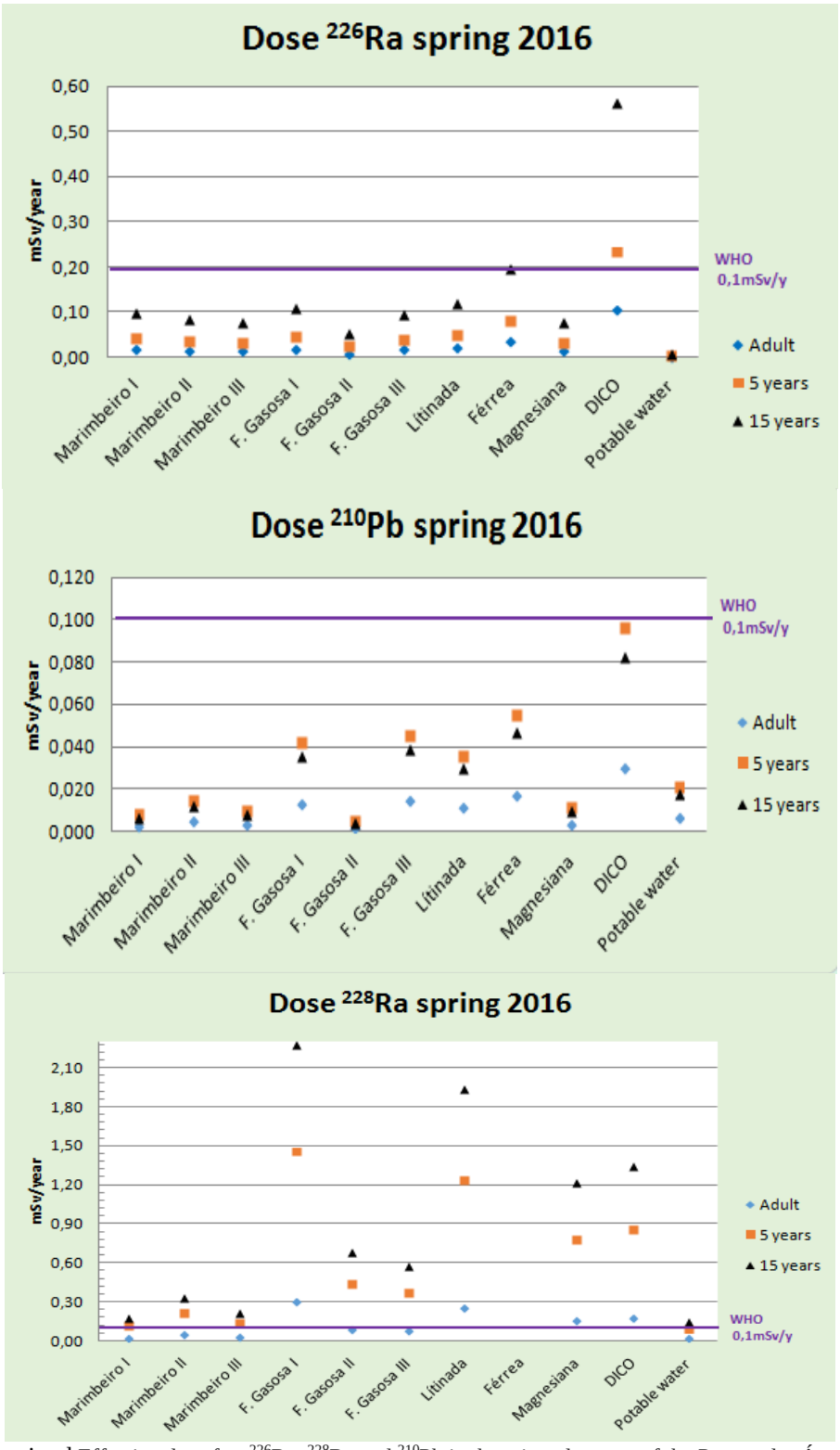

Figura 6 - Committed Effective dose for ^{226}Ra, ^{228}Ra and ^{210}Pb in the mineral waters of the Parque das Águas of Cambuquira and Marimbeiro in the spring of 2016

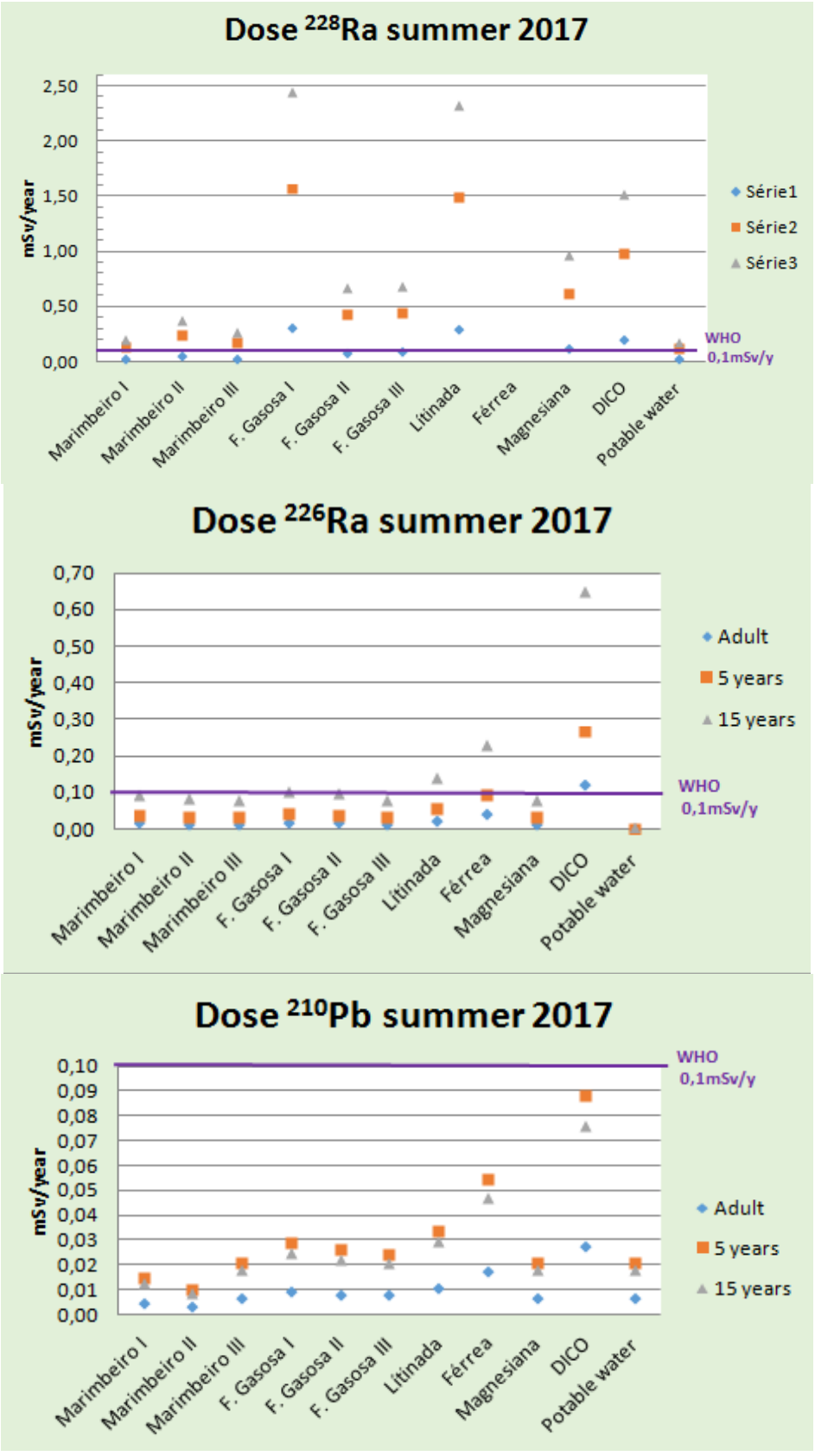

Figura 7 - Committed Effective dose for ^{226}Ra, ^{228}Ra and ^{210}Pb in the mineral waters of the Parque das Águas of Cambuquira and Marimbeiro in the Summer of 2016

In the present study the committed effective doses were also calculated for people from the public of 5 years and 15 years, since dose conversion factors are more restrictive for this age group. It was observed that for ^{228}Ra most of the spring surpassed the limit of 0.1 mSv / y , mainly for the age group of 15 years. However, it is not possible to affirm that these waters are unsuitable for consumption, since the dose limit value is always conservative and takes into account that the human consumed only the water of the Parque das águas of Cambuquira and Marimbeiro, during 1 year. It should also be remembered that the WHO [3] recommended value of 0.1 mSv / y represents 4% of the mean exposure to background natural radiation of 2.4 mSv / y [2].
For the ^{210}Pb it was possible to observe that in none of the seasons the value of 0.1 mSv / y was exceeded.

4. CONCLUSIONS

In relation to the natural radioactivity of the waters, it was possible to observe that the spring that have a ferruginous characteristic were those that presented higher concentrations of ^{226}Ra (Marimbeiro I, II, III, Laranjal or Dico and Férrea) when compared to the concentration of ^{228}Ra. It was also observed that the radionuclides ^{226}Ra and ^{228}Ra had the highest concentrations in almost of the samples in relation to ^{210}Pb and that the highest values of ^{226}Ra and ^{210}Pb were obtained in the majority of seasons for the Laranjal or Dico.
The highest committed effective doses were obtained in this work for the adult population were 1.21×10^{-01} mSv / y for ^{226}Ra referring to the spring Laranjal or Dico, in the summer collection, 3.18×10^{-01} mSv / y for ^{228}Ra referring to the spring Gasosa I also in the summer and for ^{210}Pb 3.02×10^{-02} mSv / y referring to the spring Laranjal or Dico in the spring of 2016, and also for ^{228}Ra it was possible to observe that for the adults the springs Gasosa I , Litinada, Magnesiana and Laranjal or Dico exceeded the value of 0.1 mSv / year.

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