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Natural radionuclides ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po and inorganic chemical elements determined in mineral waters from Águas de Contendas and Lambari, Brazil

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Abstract

Natural radionuclides ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po activity concentrations and the inorganic chemical composition were determined in radioactive mineral water springs from the Water Parks of Águas de Contendas and Lambari, located in the Water Circuit of the state of Minas Gerais, Brazil. Radionuclides were determined by gross alpha and beta measurements and alpha spectrometry, and the chemical elements by ICP-OES. Among the radionuclides analyzed, the highest activity concentrations obtained were for ^{226}Ra and ^{228}Ra and the chemical elements Ca, Fe and Na presented higher values of concentrations. Pearson's correlation coefficient analysis was performed to verify the correlation between natural radionuclides and the chemical elements.

Keywords Mineral water · Natural radionuclides · Inorganic chemical elements · Alpha spectrometry · ICP-OES · Water circuit

Introduction

Many studies are conducted in regions of high natural radioactivity to verify the possible biological effects on human health due to prolonged exposure to low doses of ionizing radiation, by the incorporation of natural radionuclides from the ^{238}U and ^{232}Th series, of relatively long half-lives. In some places, the levels of ^{238}U and ^{232}Th present in soil and in mineral deposits are high and, through physical and chemical dissolution and leaching mechanisms promote the passage of these radionuclides to the groundwater, from where they can emerge with a considerable activity in mineral waters [1–3].

The origin of the chemical elements present in the mineral waters is associated both to the natural environment and to the activities related to the use of the soil. In the case of the natural origin, the exogenous factors, like the geophysical and climatic aspects, along with the endogenous factors such as the lithology, the structure of the rocks and the water flow system, for example, determine the composition of the minerals that are specific to each aquifer system; this occurs because when water percolate into the soil and rock substrate to reach the aquifer, it carries the chemical components that make up these structures. Therefore, the composition of the soil and the rock substrate will determine which elements can be found in these waters and the flow of water percolation will help to determine the quantities of these elements [4]. These mineral waters may contain both essential life-sustaining chemicals, such as hydrogen, oxygen, carbon, nitrogen, calcium, phosphorus, chlorine, potassium, sulfur, magnesium, iron and zinc, as well as natural radionuclides [5].

In the same way, the natural radioactivity of mineral waters originated from underground rocks, when partially dissolved, releases some radioactive elements and gases and may be entrained by groundwater flows. However, only a few radioactive elements, such as ^{222}Rn, may be determined in the water emergence of the springs [6].

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In the aquatic environment, when the geochemistry of natural radionuclides is considered, the elements of greatest interest from the point of view of health risks are those with higher water solubility, like the isotopes of radium (Ra), radon (Rn) and uranium (U). However, elements exhibiting particle-reactive behavior, such as thorium isotopes (Th) and the radionuclides ^{210}Pb and ^{210}Po, are at lower concentration levels than the previously mentioned, except for specific cases where there is a high concentration of organic material in suspension [3].

The main source of absorption and internal exposure to natural radioactivity in humans is the diet. The ingestion of mineral water for therapeutic purposes and routine use by the local population significantly increases the probability of ingesting high concentrations of natural radionuclides, dissolved therein [7].

According to the Brazilian Law N° 7.841, of August 8, 1945 [8], mineral waters "are those from natural springs or springs artificially captured that have chemical composition or physical or physicochemical properties others than ordinary waters, with characteristics that give them a "drug action". In this same law, the chemical composition of mineral waters allows them to be classified as: oligomineral, radiferous, alkaline-bicarbonated, alkaline-earthy, sulfated, sulphurous, nitrated, chlorinated, ferruginous, radioactive, thorioactive and carbogasous.

In Brazil, radioactive mineral water springs, belonging to several hydromineral water parks, are commercially exploited, and consumed by the population that believes in the benefits of this practice. These places attract thousands of people who believe in the medicinal power of their waters. For example, in mineral water parks located in the Water Circuit of the state of Minas Gerais (Circuito das Águas do Estado de Minas Gerais), in the cities of Cambuquira, Caxambu, Conceição do Rio Verde, Lambari, and São Lourenço, the mineral waters of several springs are used both for human consumption and medicinal use, as diuretics and cathartics (with cleansing and purifying properties), facilitating hepato-biliary functions and stimulating intestinal function directly or indirectly, also with antiphlogistic (anti-inflammatory) properties [6].

Until today, the efficacy of these mineral waters used in crenological medicine has been empirical since there are no scientific studies to prove the efficacy of these radioactive mineral waters in health treatments in spas. Following the encouragement of the World Health Organization (WHO) for the integration of Traditional Medicine with Complementary/Alternative Medicine, the Brazilian Ministry of Health, through Ordinance No. 971 [9] approved the use of these Integrative Practices and Complementary by the Unified Health System (SUS) and, among them, Social Thermalism/Crenotherapy in order to prevent diseases and promote and recover health. Therefore, the importance of detailed studies of the chemical and radiochemical characterization of these mineral waters is necessary for the efficiency and safety of their use.

Hence, a research project was established with the Parks' manager from the Water Circuit of the state of Minas Gerais, in order to study their mineral waters for inorganic chemical characterization, as well as for the activity concentrations of some natural radionuclides from ^{238}U and ^{232}Th series. The objectives of this paper were to present the results of the ^{226}Ra, ^{228}Ra, ^{210}Pb, ^{210}Po activity concentrations and the elements Al, As, Ba, Ca, Cd, Cr, Fe, K, Mg, Mn, Na, Ni, Pb and Zn concentrations of the mineral waters from the Water Parks of Águas de Contendas and Lambari, as a first publication of this project.

Experimental

Study areas

The Water Parks of Águas de Contendas and Lambari belong to the cities of Conceição do Rio Verde and Lambari, in the state of Minas Gerais, respectively, Fig. 1. These cities, together with the cities of Baependi, Cambuquira, Campanha, Carmo de Minas, Caxambu, São Lourenço, Soledade de Minas and Três Corações, are part of the Water Circuit of the state of Minas Gerais. This region in Brazil concentrates the most radioactive mineral waters in the country, and these mineral waters are also classified as one of the most radioactive in the world [10–12].

The Águas de Contendas Water Park is located 7 km away from the city center of Conceição do Rio Verde and has four mineral water springs: Gasosa I, Gasosa II, Ferruginosa, and Magnesiana. The Lambari Water Park is located in downtown Lambari and has seven mineral water springs: Gasosa, Alcalina, Magnesiana, Ligeiramente Gasosa, Ferruginosa, Picante and Externa [12, 13].

Inside the parks, the springs are relatively close to one another, having different free flow rates and depths, and the water abstractions are in fountains. The climate of both cities is classified as tropical of altitude, with rainy Summers and dry Winters seasons. The springs of mineral waters are derived from a rocky substrate composed of granatiferous schists with intercalation of gneisses and muscovite quartzites, as shown in Figs. 2 and 3. The rocks are saprolized and covered by alluvial sediments superimposed by a layer of organic clay of, approximately, 5.0 m [12, 13].

Sampling

A volume of 15 L of mineral water was collected in each spring of the Water Parks in six sampling campaigns, Spring of 2016, Summer, Autumn, Winter and Spring

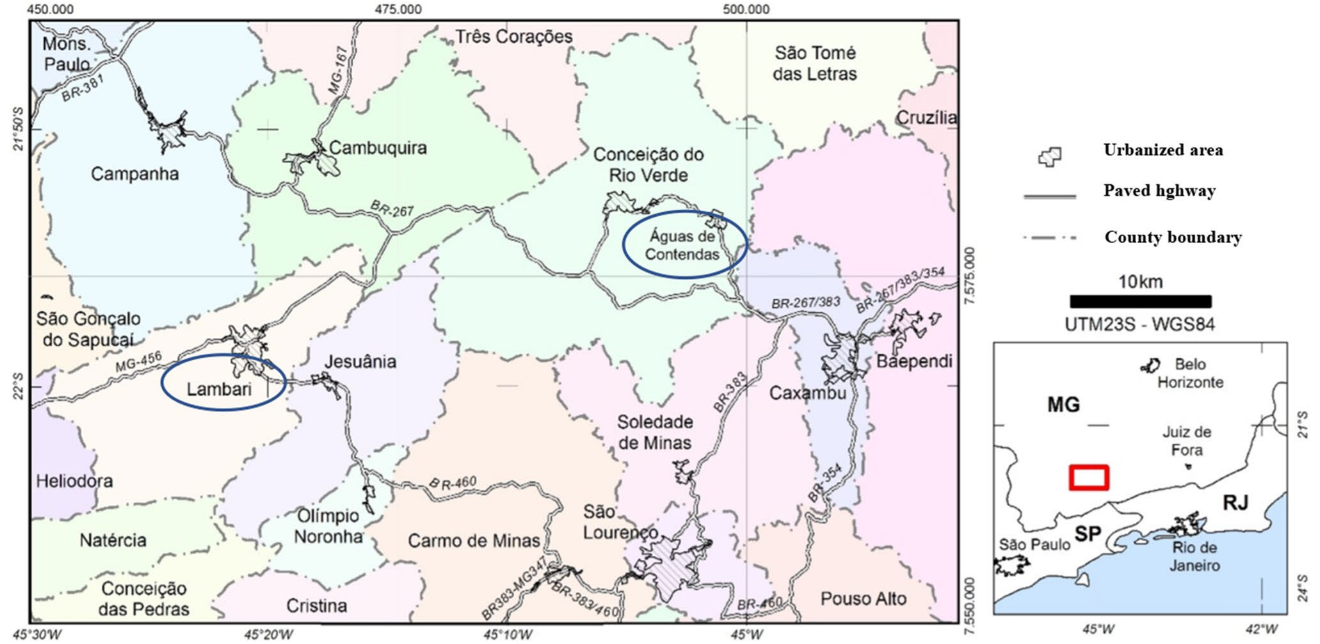

Fig. 1 Water Circuit of the state of Minas Gerais [11]

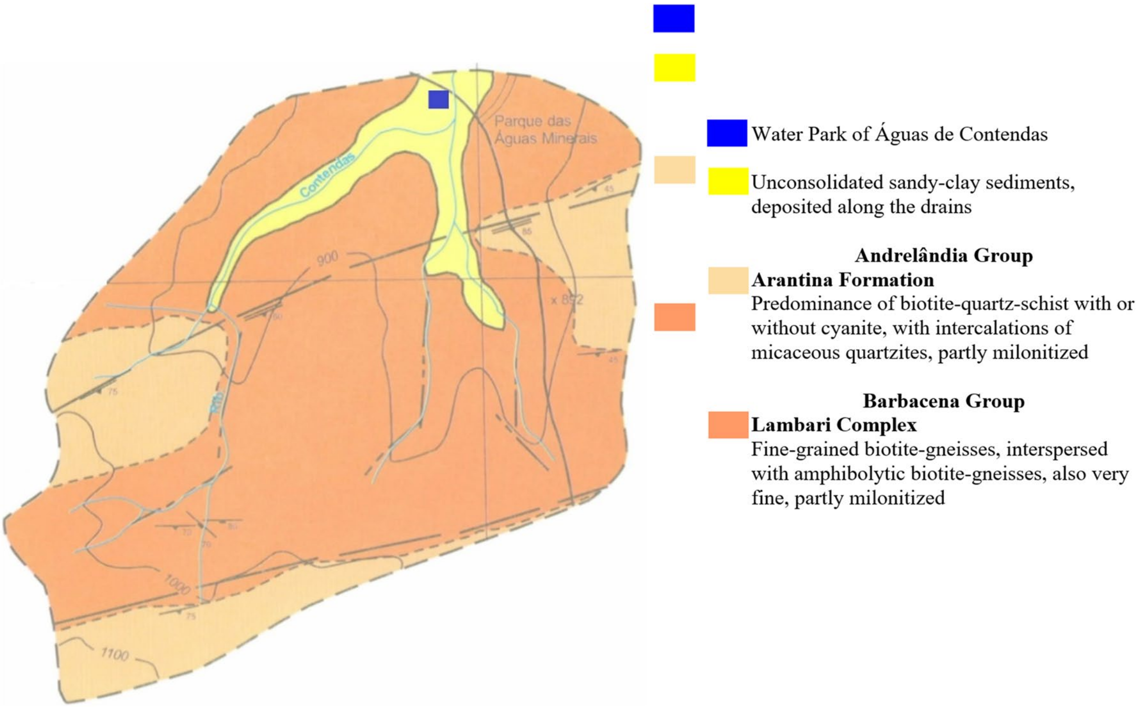

Fig. 2 Geological map of the city of Conceição do Rio Verde [12]

of 2017 and Summer of 2018; samples of drinking water were also collected in both Water Parks for comparison of the natural radionuclides and concentrations of chemical elements, determined in the mineral waters.

Right after sampling, for the preservation of the samples and, also, to avoid the adsorption of the radionuclides by the polyethylene bottles, the pH was adjusted to ≤ 2.0

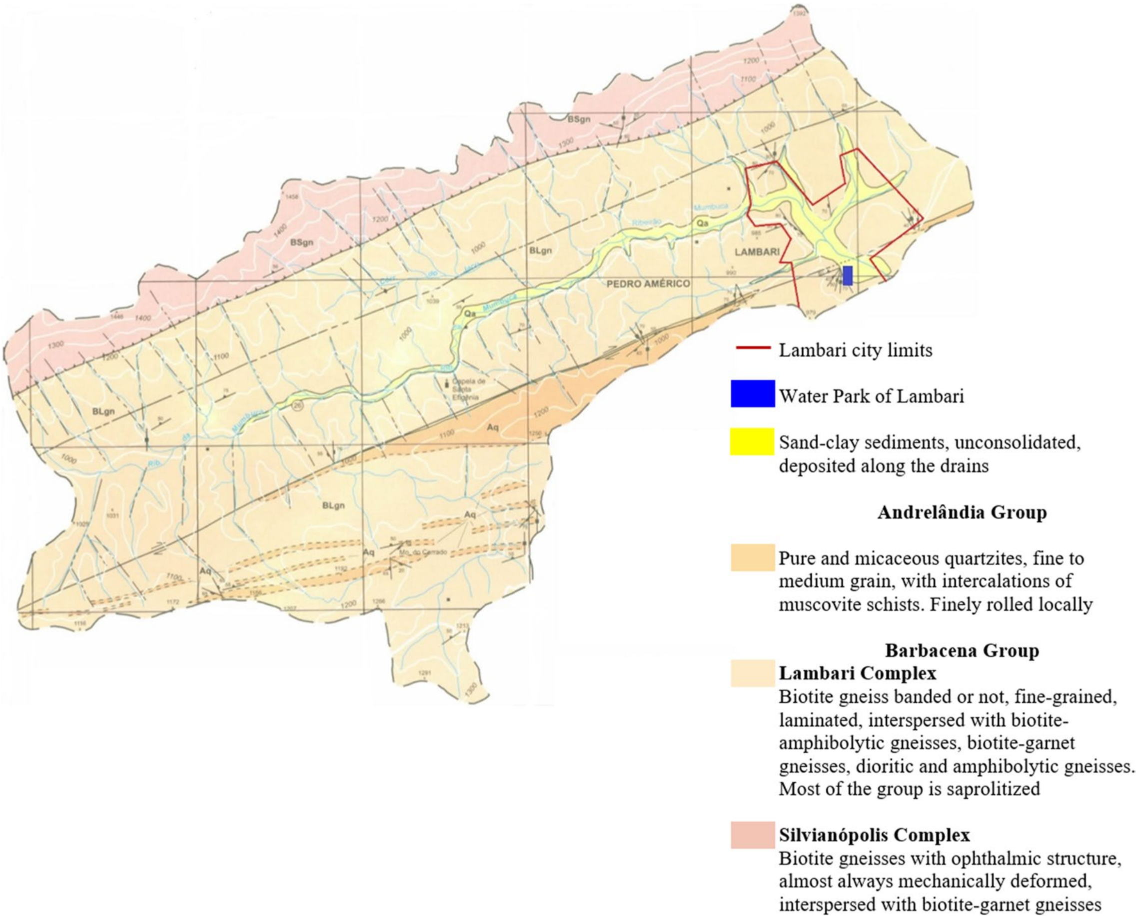

Fig. 3 Geological map of the city of Lambari [12]

with 50% HNO_3: in general, samples preserved by acid remain stable for several months [13].

Analytical methods

^{226}Ra, ^{228}Ra and ^{210}Pb activity concentration determination

The radionuclides ^{226}Ra, ^{228}Ra and ^{210}Pb were determined by gross alpha and beta measurements, after sequential radiochemical separation and measured in a low background gas flow proportional detector, Berthold model LB770-2 [14, 15].

The water samples from each spring, in duplicate, were concentrated from 2 L to 1 L in hot plate, stable Ba and Pb carriers and 50 mL of H_2SO_4 (3 mol L^{-1}) were added under heating and stirring for the precipitation of $(BaRaPb)SO_4$ with the addition of 40% NH_4OH (20–30 mL). The supernatant was discarded and, to the precipitated, 2 g of nitrile tri-acetic acid-NTA (Titriplex I), 7 mL of NaOH (6 mol L^{-1}) and 40 mL of ultrapure water were added. The solution was heated for solubilization, 5 mL of $(NH_4)_2SO_4$ (25 g L^{-1}) and glacial acetic acid (10–20 mL) were added for Ra isotopes and Ba precipitation, while ^{210}Pb remained in the supernatant, complexed with NTA.

In the precipitate containing the radium isotopes, 2 g of ethylene di-amino-tetra-acetic acid – EDTA (Triplex III), 7 mL of 40% NH_4OH and 40 mL of ultrapure water were added; the solution was heated for the precipitate solubilization. After the solubilization, $(NH_4)_2SO_4$ (25 g L^{-1}) and glacial acetic acid were added for $Ba(Ra)SO_4$ precipitation and the solution was vacuum filtered using glass fiber filter; the chemical yield of the procedure was determined gravimetrically. A low background gas flow proportional detector, Berthold model LB770-2, was used for ^{226}Ra measurement by gross alpha and ^{228}Ra by gross beta counting of the $Ba(Ra)SO_4$ precipitate, after 21 days of precipitation. This waiting time is needed for (a) Ra isotopes, ^{223}Ra and ^{224}Ra, alpha emitters with short half-life have completely decayed; (b) to achieve secular radioactive equilibrium between ^{226}Ra and its daughter ^{222}Rn (alpha emitter); (c) and, also, to achieve secular equilibrium between ^{228}Ra and its daughter ^{228}Ac (beta emitter) [15].

For ^{210}Pb determination, the supernatant containing the lead complexed with the NTA, obtained in the previous step, was used to precipitate ^{210}Pb as sulfide, with the addition of 1 mol L^{-1} Na_2S. The new precipitate was solubilized with 1 mL-2 ml of 50% HNO_3 and for the final precipitation of ^{210}Pb as $PbCrO_4$, 2.5 mL of 30% Na_2CrO_4 was added under heating. The solution obtained was vacuum filtered using glass fiber filter and the chemical yield of the procedure was determined gravimetrically. The measurement of ^{210}Pb was performed in the same low background gas flow proportional detector used for the Ra isotopes, Berthold model LB770-2, after 10 days of precipitation, time required for ^{210}Pb to achieve secular radioactive equilibrium with its daughter ^{210}Bi [15].

The measurement of the efficiency of the gas flow proportional detector for the Ra isotopes and ^{210}Pb methodology was performed using standard solutions of ^{226}Ra, ^{228}Ra and ^{210}Pb, provide by IRD-RJ, Brazil (Institute of Radioprotection and Dosimetry—Rio de Janeiro), and applying the same methodology described previously to their determination. The mean value obtained for the alpha efficiency of ^{226}Ra was 41.2%, and beta efficiency of ^{228}Ra and ^{210}Pb was 38.5% and 37.3%, respectively. A detailed explanation about the detector calibration for these radionuclides can be found in Godoy et al. [16] and Oliveira et al. [7], and about the self-attenuation of ^{226}Ra alpha counting in $Ba(Ra)SO_4$ precipitated in Godoy and Schuttelkopf [17].

The lowest limit of detection (LLD) for these radionuclides, at 95% confidence level, is 2.2 ± 0.2 mBq L^{-1} for ^{226}Ra, 3.7 ± 0.1 mBq L^{-1} for ^{228}Ra and 4.9 ± 0.4 mBq L^{-1} for ^{210}Pb; the LLD was determined using ultrapure water, with the same procedure described previously, and calculated with the Eq. (1).

$$\text{LLD} = \frac{4.66 \times S_b}{T \times Ef \times Rq \times Q} \left(\text{mBq}\,\text{L}^{-1}\right) \tag{1}$$

where LLD—lowest limit of detection, mBq L^{-1}, 4.66—Tabulated value, considering a 95% confidence level, corresponding to a pre-selected risk of a certain degree of activity in the sample, which does not exist and, vice versa, S_b—Standard deviation of the blank sample count of the process, T—Measurement time, in s, Ef—Counting system efficiency, in cps dps^{-1}, Rq—Chemical yield, Q—Sample quantity, in L

^{210}Po activity concentration determination

The radionuclide ^{210}Po was spontaneously deposited onto a cupper disc (2.0 cm diameter) following a procedure adopted by Nieri Neto and Mazzilli [18] and measured by alpha spectrometry in a Canberra detector model Alpha Analyst. The mineral water from each spring was concentrated from 1.5 L to 200 mL in hot plate, in duplicate, at controlled temperature up to 80 °C, in which ^{209}Po spike and 0.5 g to 1.0 g of ascorbic acid were added; the pH was adjusted from 1.5 to 2.0, with 40% NH_4OH. The sample was heated under stirring at controlled temperature up to 80 °C for the deposition of both Po, for 4 h; the counting time was 250,000 s [19, 20]. The lowest limit of detection (LLD) for ^{210}Po determination, using this methodology, is 3.3 ± 0.4 mBq L^{-1}. The ^{210}Po LLD determination was also performed using ultrapure pure water and with the same procedure described above and calculated using Eq. (1). The counting efficiency of the alpha spectrometer was performed using an electrodeposited (2.0 cm diameter) and calibrated ^{241}Am source, also provided by IRD-RJ, and the mean value was 45.7%.

The methodologies used for ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po determinations were validated using the reference material Natural and Artificial Radionuclides in Sediment from the Irish Sea 385—IAEA. The mean values obtained for the radionuclides studied, in 10 replicates, as well as, the relative standard deviation and relative errors are presented in Table 1; the results obtained are in good agreement with the certified values.

Inorganic chemical characterization

The elements Al, As, Ba, Ca, Cd, Cr, Fe, K, Mg, Mn, Na, Ni, Pb and Zn were determined by the Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) with argon plasma, Spectro model Arcos, in the Summer and Winter sampling of 2017; Ferruginosa spring, from the Water Park of Lambari was only analyzed in Summer of 2018 [21]. For this analysis, 200 mL of mineral water from each spring, acidified with 50% HNO_3, was concentrated to

Table 1 ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po methodologies validation using the reference material Natural and Artificial Radionuclides in Sediment from the Irish Sea 385—IAEA

Radionuclide	M ± SD (Bq kg^{-1})	Certified value (Bq kg^{-1})	Confidence interval	Relative standard deviation (%)	Relative error (%)
^{226}Ra	20.5 ± 3.9	21.9	21.6–22.4	19	6.4
^{228}Ra	35.8 ± 4.8	32.0	31.3–32.5	13	12
^{210}Pb	32.5 ± 6.5	32.9	31.2–35.3	20	1.2
^{210}Po	34.9 ± 0.8	32.9	31.2–35.3	2.3	6.1

Table 2 Minimum limits of quantification (MLQ) of the chemical elements determined by ICP-OES

Element (mg/kg)	MLQ
Al	0.05
As	0.005
Ba	0.05
Ca	10.143
Cd	0.043
Cr	0.01
Fe	0.05
K	0.055
Mg	0.201
Mn	0.01
Na[a]	6.756
Ni	0.01
Pb	0.008
Zn	0.021

[a] in mg/L

20 mL and analyzed into the ICP-OES, in three replicates per sample. The minimum limits of quantification for this methodology are shown in Table 2 and the expanded uncertainty associated with this methodology ranged from 3 to 18% [22].

Results and discussion

^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po activity concentrations

The activity concentrations of ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po obtained for the mineral water samples, are presented in Table 3, for the six sampling campaigns; in most samples the radionuclides with the highest concentrations were ^{226}Ra and ^{228}Ra.

At the Water Park of Águas de Contendas, the highest activity concentration values obtained were for ^{226}Ra, 77 ± 5 mBq L^{-1}; for ^{228}Ra, 202 ± 2 mBq L^{-1}; for ^{210}Pb, 51 ± 5 mBq L^{-1}, and ^{210}Po, 16 ± 1 mBq L^{-1}. At Parque das Águas of Lambari, the highest activity concentration values were for ^{226}Ra, 250 ± 3 mBq L^{-1}; for ^{228}Ra, 244 ± 4 mBq L^{-1}; for ^{210}Pb, 57 ± 3 mBq L^{-1} and for ^{210}Po, 13 ± 1 mBq L^{-1}.

The drinking water samples collected in the Water Park of Lambari presented activity concentrations that ranged for ^{226}Ra from < LLD (2.2 ± 0.2 mBq L^{-1}) to 5.9 ± 0.2 mBq L^{-1}; for ^{228}Ra from 19.3 ± 0.4 mBq L^{-1} to 87 ± 1 mBq L^{-1}; for ^{210}Pb from < LLD (4.9 ± 0.4 mBq L^{-1}) and for ^{210}Po all the activity concentrations presented values < LLD (3.3 ± 0.4 mBq L^{-1}).

At Water Park of Águas de Contendas the collected drinking water samples presented values of activity concentrations for ^{210}Pb and ^{210}Po higher or of the same magnitude than the mineral waters. Thus, one sample of drinking water was collected 7 km away from the park, in the city of Conceição do Rio Verde; the results obtained for this sample presented values below the DL for ^{226}Ra and ^{210}Po, and lower values for ^{228}Ra and ^{210}Pb than the mineral water from the springs. These results indicate an influence of the local geology on the radionuclide activity concentrations in the drinking water samples collected near the Water Park, although this drinking water has been treated for human consumption, proving that the water treatment process, to make it drinkable, removes much of the natural radioactivity present.

Figure 4 shows the variation of the activity concentrations for the analyzed radionuclides in both Water Parks and in all the springs and seasons. It may be observed that, in general, the concentrations obtained in the Water Park of Lambari were higher than those obtained in Águas de Contendas, the radionuclides ^{226}Ra, ^{228}Ra and ^{210}Pb presented the highest values in Lambari and only ^{210}Po presented higher values in Águas de Contendas. It is also possible to observe that the highest and the lowest concentrations of the radionuclides studied oscillated among the six seasons, without a standard concentration behavior. Despite this, as the variations in the activity concentration are small, seasonality is barely noticeable between the seasons analyzed.

According to Ordinance 2914 of the Brazilian Ministry of Health, the maximum value allowed (MVA) for the natural radionuclide ^{226}Ra is 1 Bq L^{-1} and, for ^{228}Ra, is 0.1 Bq L^{-1} in drinking water [23, 24]. The results obtained in the present work for ^{226}Ra are below the MVA for all mineral water samples, while for ^{228}Ra in some seasons, the concentrations determined exceeded the MVA, such as Gasosa I and Gasosa II springs, from the Water Park of Águas de Contendas and for all the springs of the Water Park of Lambari.

Few studies may be found in the literature determining the activity concentrations of the studied radionuclides, in the Water Parks from Minas Gerais, Brazil. At the present date, there are no studies evaluating the activity concentration of ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po in the Water Park of Águas de Contendas and the radionuclides ^{210}Pb and ^{210}Po, for the Water Park of Lambari. Therefore, the results showed in the present work are inedited for the mineral waters analyzed.

One of the studies that analyzed the mineral waters from the Water Park of Lambari was performed by Bonotto [25], determining the activity concentrations of ^{226}Ra and ^{228}Ra by alpha spectrometry and obtaining a range from 41.8 to 448.9 mBq L^{-1} and from 106.6 to 237.6 mBq L^{-1}, respectively. The values achieved by the author were higher than those determined in the present work, at Parque das Águas of Lambari.

However, the activity concentrations results determined in the present study were compared with works that analyzed mineral waters from other Water Parks of the Water Circuit of Minas Gerais, for example, Meneghini et al. [26],

Table 3 ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po activity concentrations in mineral water samples from Water Park of Águas de Contendas and Lambari

Water park		Spring 2016 (mBq/L ± SD)				Summer 2017 (mBq/L ± SD)				Autumn 2017 (mBq/L ± SD)			
	Samples	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po
Águas de Contendas	Ferruginosa	24 ± 1	57 ± 2	15 ± 1	11 ± 1	22 ± 1	67 ± 1	15 ± 1	11.4 ± 0.1	34 ± 3	< DL	18.6 ± 0.5	< DL
	Magnesiana	20 ± 3	45 ± 3	27 ± 1	14 ± 1	***	***	***	***	20 ± 1	< DL	30 ± 2	9 ± 1
	Gasosa I	68 ± 2	129 ± 5	11 ± 1	9 ± 1	70 ± 5	123 ± 5	15 ± 2	6.2 ± 0.3	35 ± 3	114 ± 5	24 ± 2	10 ± 1
	Gasosa II	77 ± 5	127 ± 5	19 ± 2	6.5 ± 0.4	45 ± 1	106 ± 6	12 ± 1	6.4 ± 0.6	60 ± 1	12 ± 1	26 ± 2	5.0 ± 0.1
	D. Water—AC	16 ± 2	< DL	< DL	4.0 ± 0.4	12.9 ± 0.5	59 ± 4	13 ± 1	13.7 ± 0.3	21 ± 1	< DL	33 ± 2	18 ± 1
	D. water—CRV	*	*	*	*	*	*	*	*	*	*	*	*
Lambari	Alcalina	151 ± 3	72 ± 5	25.0 ± 0.3	7.6 ± 0.7	171 ± 5	110 ± 6	14 ± 1	5.3 ± 0.5	162 ± 2	119 ± 3	19.8 ± 1.8	< DL
	Magnesiana	141 ± 4	135 ± 5	34 ± 2	10 ± 1.0	177 ± 2	72 ± 1	36 ± 2	12.7 ± 0.2	149 ± 3	48 ± 5	38.2 ± 2.0	13 ± 1
	Gasosa	163 ± 4	44 ± 2	26 ± 1	7.5 ± 0.7	155 ± 3	127 ± 5	27.2 ± 0.4	8.9 ± 0.1	189 ± 9	34 ± 2	17.7 ± 0.6	9.1 ± 0.7
	L. Gasosa	118 ± 3	37 ± 2	20 ± 2	5.5 ± 0.5	87 ± 3	96 ± 5	32 ± 3	8.0 ± 0.7	99 ± 3	78 ± 5	14.5 ± 1.4	9.7 ± 0.6
	Picante	101 ± 3	91 ± 3	9.0 ± 0.9	11 ± 1	118 ± 3	36 ± 4	17.6 ± 0.2	6.3 ± 0.5	104 ± 7	29 ± 5	14.3 ± 1.5	5.0 ± 0.2
	Drinking Water	5.0 ± 0.1	87 ± 1	8.4 ± 0.8	< DL	5.2 ± 0.4	52 ± 6	< DL	< DL	5.9 ± 0.2	50 ± 3	< DL	< DL
	Externa	*	*	*	*	*	*	*	*	136 ± 3	59 ± 5	26 ± 1	5.4 ± 0.5
	Ferruginosa	**	**	***	***	**	**	***	***	**	**	***	***
Water Park	Samples	Winter 2017 (mBq/L ± SD)				Spring 2017 (mBq/L ± SD)				Summer 2018 (mBq/L ± SD)			
		^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po
Águas de Contendas	Ferruginosa	34 ± 1	13 ± 1	16.2 ± 0.7	10 ± 1	**	**	***	***	7.7 ± 0.3	61.2 ± 0.3	51 ± 5	12.7 ± 0.2
	Magnesiana	9.8 ± 0.3	26 ± 1	20 ± 2	15.7 ± 0.5	**	**	***	***	7.9 ± 0.4	66 ± 3	48 ± 5	12.8 ± 0.6
	Gasosa I	70 ± 2.0	86 ± 2	5.5 ± 0.7	13.7 ± 0.4	68.4 ± 0.5	< DL	7.8 ± 0.9	7.6 ± 0.4	30.2 ± 0.6	202 ± 2	24 ± 2	10.7 ± 0.7
	Gasosa II	61 ± 2.0	52 ± 3	7.8 ± 0.8	5.5 ± 0.2	**	**	***	***	26.0 ± 0.4	135 ± 2	14 ± 1	12.8 ± 0.2
	D. Water - AC	17.7 ± 0.5	22 ± 1	19 ± 2.0	7.3 ± 0.2	10.8 ± 0.2	< DL	19.6 ± 0.2	17.7 ± 0.4	8.0 ± 0.6	80.2 ± 0.1	16 ± 2	24.3 ± 0.2
	D. Water - CRV	*	*	*	*	*	*	*	*	< DL	64 ± 3	6.9 ± 0.2	< DL
Lambari	Alcalina	223 ± 3	< DL	36 ± 3	9.1 ± 0.8	156 ± 5	79 ± 4	10.9 ± 0.8	9.7 ± 0.2	82 ± 3	244 ± 4	31 ± 3	3.8 ± 0.4
	Magnesiana	208 ± 1	< DL	37.2 ± 0.4	10.2 ± 0.9	160 ± 3	35 ± 2	26 ± 2.2	13 ± 1	83 ± 6	202 ± 4	38 ± 0.6	13.4 ± 0.1
	Gasosa	202 ± 3	165 ± 5	57 ± 3	6.8 ± 0.6	155 ± 7	63 ± 3	15 ± 1	11.4 ± 0.4	79 ± 3	238 ± 5	25 ± 2	6.3 ± 0.5
	L. Gasosa	137 ± 7	19.3 ± 0.5	35 ± 4	8.8 ± 0.7	104 ± 3	44 ± 2	< DL	13.4 ± 0.3	54 ± 2	177 ± 7	34 ± 3	< DL
	Picante	139 ± 7	< DL	15.2 ± 0.3	8.8 ± 0.7	102 ± 3	13 ± 1	< DL	11.6 ± 0.6	41 ± 2	149 ± 5	9.9 ± 0.9	7.0 ± 0.8
	D. Water	3.6 ± 0.4	19.2 ± 0.4	< DL	< DL	3.6 ± 0.7	43 ± 2	< DL	< DL	< DL	48 ± 2	5.6 ± 0.5	< DL
	Externa	250 ± 3	< DL	23 ± 1	5.0 ± 0.1	164 ± 3	70 ± 4	13 ± 1	8.7 ± 0.7	74 ± 4	232 ± 5	21.1 ± 0.2	9.2 ± 0.9
	Ferruginosa	**	**	***	***	**	**	***	***	56 ± 3	127 ± 5	11 ± 1	7.5 ± 0.5

*Uncollected sample ** Park under renovation *** Dry spring ^{226}Ra DL = 2.2 ± 0.2 mBq/L ^{228}Ra DL = 3.7 ± 0.1 mBq/L ^{210}Pb DL = 4.9 ± 0.4 mBq/L ^{210}Po DL = 3.3 ± 0.4 mBq/L SD = Standard Deviation

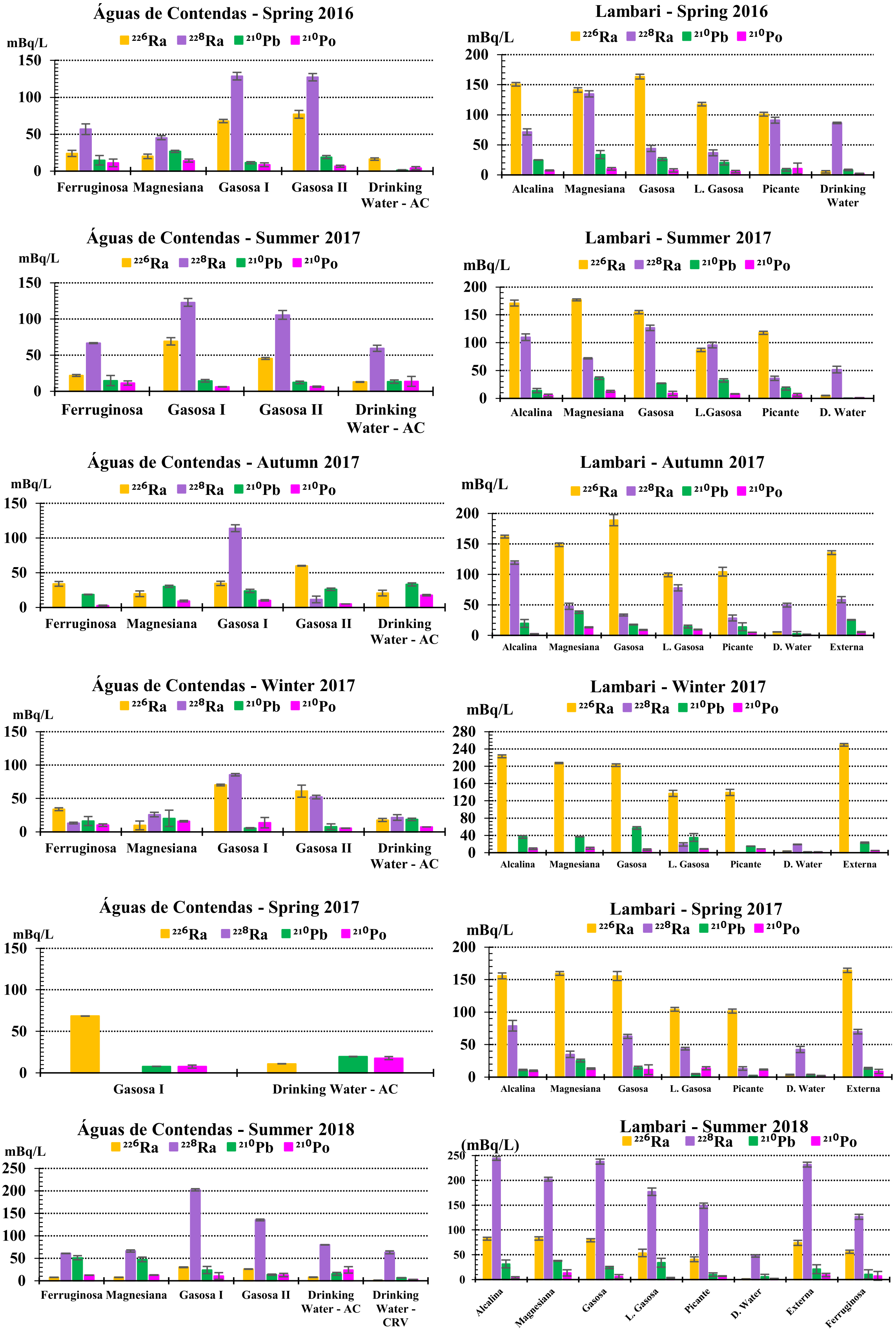

Fig. 4 ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po activity concentrations in the mineral waters of Águas de Contendas and Lambari Water Parks

who determined the activity concentrations of ^{226}Ra, ^{228}Ra and ^{210}Pb at Water Park of Caxambu, and Santos et al. [27], who analyzed ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po at Water Park of Cambuquira and Marimbeiro. The results obtained by these authors are higher than those in the present work, despite the proximity of the parks; they are part of distinct geological groups and therefore confer different lithologies through which the waters percolate enriching these mineral waters.

Nevertheless, to compare the results of the present study with data from the literature outside Brazil is slightly inaccurate and difficult, because one of the most important factors that influence radionuclide concentrations is the geology of the studied area. As examples, it may be mentioned the work performed by Wallner et al. [28], who analyzed the natural radionuclides ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po in Austrian mineral waters, obtaining the majority of activity concentrations lower than the present study for ^{210}Pb and ^{210}Po, however for ^{226}Ra and ^{228}Ra, most values were superior only to those found in Águas de Contendas samples. Jobbágy et al. [29], analyzed, by a radiochemical method, ^{226}Ra and ^{210}Po in mineral water samples from Belgium, France, Italy and Poland, obtaining activity concentration values lower than those determined by the present study for ^{210}Po and for ^{226}Ra, in some samples from Belgium and France, and Erden et al. [30] analyzed the concentration of ^{226}Ra in mineral waters from Turkey and obtained higher values than those determined by the present study.

Inorganic chemical characterization

Table 4 shows the concentration of the inorganic chemical elements Al, As, Ba, Ca, Cd, Cr, Fe, K, Mg, Mn, Na, Ni, Pb and Zn, determined by ICP-OES in the mineral water samples from both Water Parks, for the samples collected in the Summer (Sum) and Winter of 2017 (Win). These chemical elements were analyzed in the water samples, since they are one of the parameters demanded by the Brazilian Ministry of Health [23] and by the Conselho Nacional do Meio Ambiente—CONAMA (National Council for the Environment) [24] to classify whether a sample of water is potable; this table also presents the Maximum Permitted Value (MPV) for these elements, assigned by these Governmental Organizations, as well as the Total Hardness of the Water, which is another parameter for water potability.

The concentration results of the elements As, Cd, Cr and Ni presented in this work are unpublished and Cd element presented values below the Minimum Limit of Quantification in all samples analyzed.

For both Water Parks, in general, the chemical elements that presented the highest concentrations were the major elements, Ca, Fe and Na; these elements come from the rocky substrate by which water percolates and is stored [31–34], in most of the springs. The highest concentrations obtained in the Water Park of Águas de Contendas were 139.43 ± 0.03 mg L^{-1} for Na, 36.440 ± 0.001 mg L^{-1} for Fe and 34.100 ± 0.001 mg L^{-1} for Ca; at Lambari, the highest concentrations were 12.63 ± 0.02 mg L^{-1} for Ca and 11.29 ± 0.07 mg L^{-1} for Na. In the same way, the variations in the concentration of these inorganic chemical elements are small as those observed in the radionuclide activity concentrations, thus, seasonality is also barely noticeable between the seasons analyzed.

The drinking water from the Water Parks of Águas de Contendas and Lambari presented the highest concentrations for Ca, 16.98 ± 0.03 mg L^{-1} and 8.45 ± 0.02 mg L^{-1}, respectively, lower than the mineral waters. The drinking water sample of Lambari, also, presented concentrations of Fe above the MPV [23, 24]; the presence of this element in higher concentrations may be due to the oxidation of the piping system that distributes the water.

The Total Hardness of Water—THW, expressed in mg L^{-1}, is the sum of the temporary and permanent hardness and it is defined by the total concentration of Ca and Mg. It may vary by the rock and soil composition of the studied area. In the present work, it was calculated according to Hypolito et al. [35], by summing the concentrations of Ca and Mg elements multiplied by conversion factors, Eq. (2):

$$\mathrm{THW} = \left(\mathrm{Ca}_{\mathrm{con}}.2.5\right) + \left(\mathrm{Mg}_{\mathrm{conc}}.4.12\right)\left(\mathrm{mg\ L}^{-1}\right) \tag{2}$$

where THW—total hardness of water, mg L^{-1}, Ca_{con} and Mg_{conc}—Ca and Mg concentration. mg L^{-1}, respectively.

THW results classify the waters as soft, less than 50 mg L^{-1}; slightly hard, up to 100 mg L^{-1}; moderately hard, up to 200 mg L^{-1}, and values greater than 200 mg L^{-1} as very hard; THW variation in surface water and groundwater ranges from 10 to 300 mg L^{-1} [4].

At the Water Park of Águas de Contendas, the values obtained varied from 6.804 ± 0.015–94.64 ± 0.08 mg L^{-1}, in the Summer of 2017. At the Water Park of Lambari, the values ranged from 10.15 ± 0.10 to 41.13 ± 0.03 mg L^{-1}, also in the Summer of 2017. These results of THW classify the springs of both Water Parks as soft, THW < 50 mg L^{-1} and were below the MPV for drinking water, stipulated by the Brazilian Ministry of Health (500 mg L^{-1}) [23].

The mineral waters from both Water Parks were analyzed by the Company of Research of Mineral Resources [11] that determined the elements Al, As, Ba, Ca, Fe, K, Mg, Mn, Na and Zn, in both Water Parks, and the Company of Economic Development of Minas Gerais [12] that analyzed the mineral waters from Águas de Contendas, determining the elements Ba Ca. K, Mg and Na. The chemical elements analyzed by these two companies presented lower values than those obtained in the present work in most of the water samples; the differences among

Table 4 Inorganic chemical elements determined by ICP-OES in mineral water samples from the Water Parks of Águas de Contendas and Lambari in Summer and Winter 2017

		Al	As	Ba	Ca[1]	Cr	Fe[1]	K[1]	Mg[1]	Mn	Na[1]	Ni	Pb	Zn	THW[1]
Águas de Contendas—μg/L															
Ferruginosa	Sum	242	2.91	65.0	8.10	2.75	16.0	3.60	2.21	170	3.62	186	22.7	30.7	29.4
	Win	183	4.50	52.0	10.1	2.70	17.0	3.60	2.28	196	4.27	181	25.0	34.5	34.6
Magnesiana	Sum	182	0.55	16.6	1.85	0.85	0.49	1.67	0.53	11.6	1.66	49.8	9.74	122	6.80
	Win	300	1.70	35.4	3.91	2.81	1.24	3.25	1.03	24.5	2.75	95.0	15.7	182	14.0
Gasosa I	Sum	118	2.70	130	11.0	2.20	0.17	4.66	2.31	164	39.7	122	18.2	34.5	37.0
	Win	196	3.40	183	15.2	3.40	0.18	7.65	2.32	254	74.7	303	27.0	31.7	47.6
Gasosa II	Sum	220	6.50	110	34.1	2.60	36.4	10.2	2.28	648	136	461	23.0	39.0	94.6
	Win	279	8.00	131	31.3	3.32	34.8	9.58	2.31	590	139	495	31.3	46.0	87.9
D. W.—AC	Sum	43.5	2.30	17.3	14.1	2.67	0.03	3.25	2.29	0.93	8.02	55.8	15.4	96.0	44.7
	Win	62.0	2.83	16.9	17.0	3.13	0.03	3.01	2.32	0.81	9.36	64.8	14.9	212	52.0
D. W.—CRV	Sum	330	< MLQ	28.6	5.48	1.11	0.13	2.89	1.20	6.23	3.94	16.7	9.90	57.6	18.6
Lambari—μg/L															
Alcalina	Sum	146	1.60	183	9.50	2.61	39.1	3.89	2.29	88.4	7.90	26.4	7.80	19.4	33.2
	Win	130	4.59	194	9.60	2.21	22.9	4.02	2.29	92.0	8.09	27.6	14.3	10.4	33.4
Magnesiana	Sum	102	1.80	134	8.70	1.61	20.9	3.21	2.15	72.0	7.70	19.0	7.20	7.96	30.6
	Win	125	2.10	175	10.3	2.27	26.1	3.79	2.26	94.1	10.2	24.6	9.60	15.1	35.0
Gasosa	Sum	131	1.92	191	9.26	2.43	93.9	3.92	2.29	100	8.47	24.7	9.60	12.4	32.6
	Win	179	2.10	190	9.45	3.00	76.1	4.12	2.30	99.0	8.81	26.3	9.90	23.4	33.1
Ligeiramente Gasosa	Sum	127	1.80	158	7.34	2.67	203	3.48	2.11	69.7	11.3	21.9	11.1	10.1	27.0
	Win	272	1.38	118	6.02	2.37	89.0	3.99	1.81	57.0	8.61	19.0	8.90	15.2	22.5
Picante	Sum	346	1.98	228	12.6	2.49	90.3	4.96	2.32	116	7.40	43.0	15.2	22.8	41.1
Drinking Water	Sum	111	2.70	35.5	8.09	1.74	226	1.40	1.85	19.8	4.28	23.2	10.1	7.44	27.8
	Win	674	1.50	49.5	8.45	2.63	1030	1.50	1.88	325	5.00	28.5	8.40	21.1	28.9
Externa	Sum	43.7	0.55	50.5	2.60	0.79	9.97	1.17	0.89	25.7	2.41	7.55	3.00	6.30	10.2
	Win	137	1.56	193	9.64	2.11	27.8	3.99	2.29	98.0	9.40	28.8	7.10	11.8	33.5
Ferruginosa	Sum	107	0.55	40.4	2.56	0.59	81.8	1.09	0.91	24.1	1.27	8.45	3.00	6.13	10.1
MPV (mg/L) Drinking Water[a]		0.2	0.01	0.7	[b]	0.05	0.3	[b]	[b]	0.1	200	0.07[c]	0.01	5	500[d]

MLQ minimum limit of quantification, *MPV* maximum permitted value, *THW* total hardiness of water, *AC* Águas de contendas, *CRV* conceição do Rio verde sum, *summer win* winter

[a] Values assigned by CONAMA and Brazilian Ministry of Health [23, 24]

[b] MPV not determined

[c] Value of 0.02 assigned by CONAMA [24]

[d] MPV assigned only by Brazilian Ministry of Health [23]

[1] Unit of concentration of the elements in mg/L

the results may be due to the sample collection period and, the analysis methodology used by both companies. It should, also, be emphasized the importance of geo-climatic and hydrogeochemical factors for the concentration of these elements over time.

Figure 5 shows the variation in the concentration of the inorganic chemical elements; it may be observed that, in Summer, the mineral waters from Águas de Contendas presented the highest concentrations, when compared with Lambari, while in Winter, opposite concentrations occurred.

At the Water Park of Águas de Contendas it was possible to observe a pattern of concentration between the two seasons; Mn, Na, Ni and Zn elements presented the highest concentrations in both seasons and Ca, K, Mg and Na elements, in Lambari.

Statistical analyzes

Pearson's correlation coefficient analysis was performed with the mean activity concentrations of ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po, Fig. 6. For each park, separately, it is possible to observe that at the Water Park of Águas de Contendas only ^{226}Ra and ^{210}Pb presented a strong correlation, inversely proportional ($r = -0.922$) and, in Lambari, between ^{226}Ra and

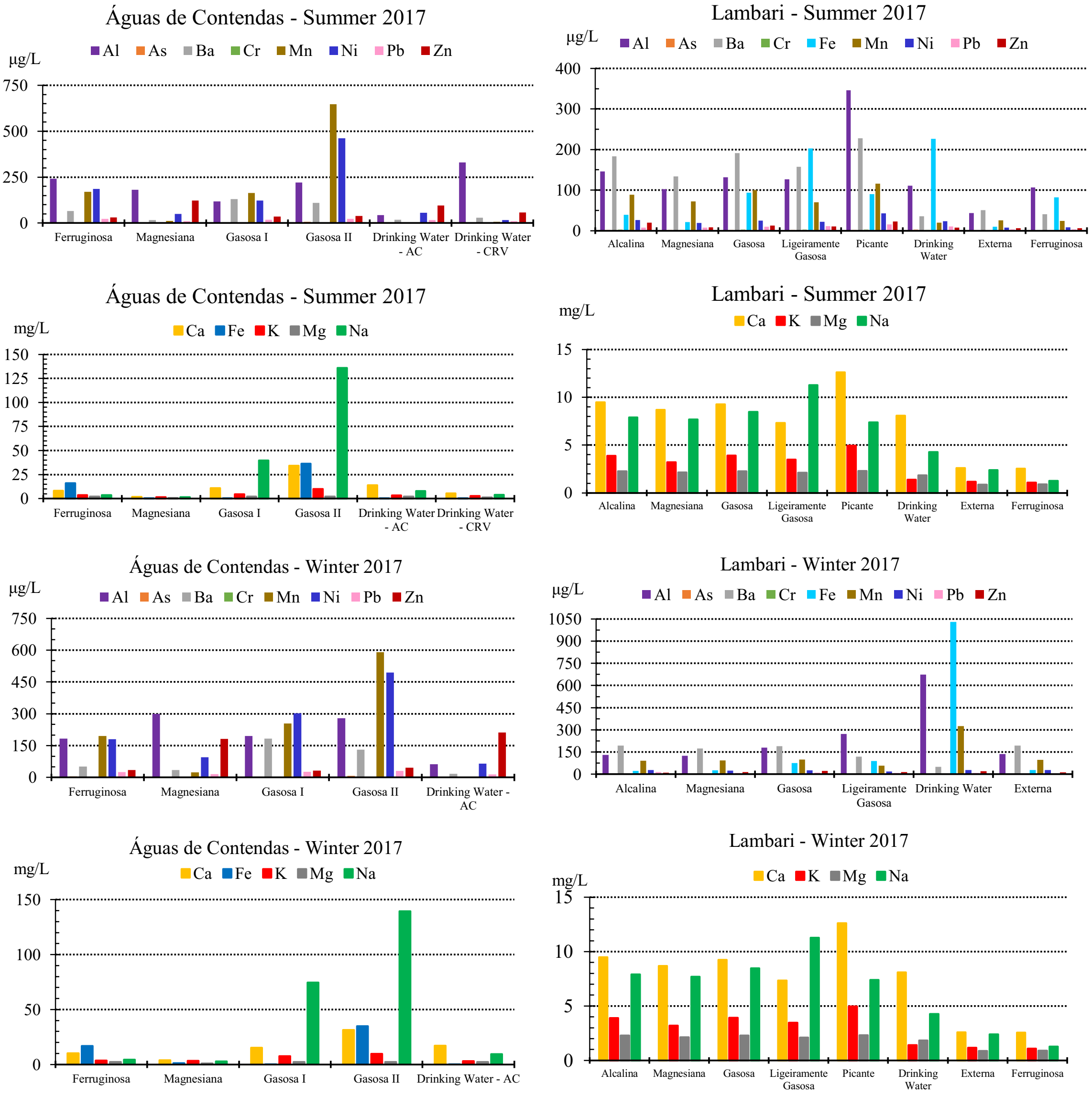

Fig. 5 Inorganic chemical element concentrations determined by ICP-OES in water samples from the Water Parks of Águas de Contendas and Lambari

Fig. 6 Pearson's correlation coefficient analysis, with the mean activity concentrations of ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po determined in the mineral waters

		Águas de Contendas				Lambari			
		^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po
Águas de Contendas	^{226}Ra	1							
	^{228}Ra	-0.696	1						
	^{210}Pb	-0.922	0.585	1					
	^{210}Po	-0.577	0.568	0.424	1				
Lambari	^{226}Ra	0.566	-0.563	-0.766	-0.269	1			
	^{228}Ra	-0.755	0.751	0.817	0.529	-0.927	1		
	^{210}Pb	-0.492	0.398	0.140	0.568	0.366	-0.014	1	
	^{210}Po	0.903	-0.685	-0.749	-0.4598	0.256	-0.472	-0.691	1

		Águas de Contendas													Lambari												
		Al	As	Ba	Ca	Cr	Fe	K	Mg	Mn	Na	Ni	Pb	Zn	Al	As	Ba	Ca	Cr	Fe	K	Mg	Mn	Na	Ni	Pb	Zn
Águas de Contendas	Al	1																									
	As	0.293	1																								
	Ba	-0.592	0.507	1																							
	Ca	0.263	0.966	0.604	1																						
	Cr	-0.303	0.804	0.774	0.740	1																					
	Fe	0.576	0.936	0.171	0.856	0.604	1																				
	K	0.096	0.908	0.742	0.979	0.759	0.734	1																			
	Mg	-0.426	0.695	0.754	0.610	0.984	0.488	0.639	1																		
	Mn	0.291	0.985	0.563	0.996	0.757	0.895	0.960	0.632	1																	
	Na	0.187	0.879	0.681	0.972	0.660	0.726	0.990	0.523	0.948	1																
	Ni	0.262	0.982	0.590	0.997	0.769	0.881	0.968	0.646	0.999	0.954	1															
	Pb	-0.130	0.880	0.667	0.794	0.983	0.734	0.778	0.950	0.822	0.687	0.828	1														
	Zn	0.466	-0.635	-0.715	-0.534	-0.964	-0.436	-0.562	-0.995	-0.561	-0.439	-0.574	-0.925	1													
Lambari	Al	0.102	0.730	0.715	0.882	0.526	0.541	0.939	0.389	0.836	0.967	0.846	0.523	-0.300	1												
	As	-0.250	-0.363	0.279	-0.110	-0.381	-0.532	0.045	-0.420	-0.198	0.124	-0.180	-0.486	0.461	0.371	1											
	Ba	-0.703	-0.360	0.563	-0.155	-0.068	-0.643	0.047	-0.039	-0.235	0.056	-0.206	-0.235	0.053	0.277	0.862	1										
	Ca	-0.370	-0.694	0.046	-0.486	-0.591	-0.814	-0.334	-0.570	-0.562	-0.268	-0.545	-0.713	0.575	-0.016	0.921	0.843	1									
	Cr	-0.327	0.338	0.829	0.550	0.367	0.045	0.700	0.288	0.476	0.720	0.499	0.273	-0.219	0.857	0.706	0.731	0.423	1								
	Fe	0.091	0.827	0.746	0.941	0.652	0.639	0.983	0.523	0.908	0.993	0.917	0.658	-0.438	0.986	0.220	0.176	-0.168	0.798	1							
	K	-0.252	0.105	0.640	0.353	0.072	-0.144	0.508	-0.007	0.268	0.561	0.289	-0.023	0.075	0.750	0.879	0.805	0.642	0.954	0.646	1						
	Mg	-0.306	-0.357	0.325	-0.105	-0.340	-0.544	0.058	-0.373	-0.194	0.130	-0.174	-0.454	0.412	0.376	0.998	0.891	0.920	0.728	0.229	0.889	1					
	Mn	-0.707	-0.574	0.370	-0.389	-0.256	-0.808	-0.196	-0.200	-0.463	-0.183	-0.437	-0.424	0.195	0.047	0.840	0.970	0.911	0.550	-0.064	0.670	0.865	1				
	Na	0.278	0.294	0.346	0.521	-0.053	0.191	0.585	-0.199	0.453	0.683	0.458	-0.054	0.288	0.820	0.723	0.417	0.415	0.785	0.716	0.859	0.703	0.274	1			
	Ni	-0.656	-0.302	0.588	-0.086	-0.049	-0.587	0.114	-0.035	-0.169	0.129	-0.140	-0.210	0.056	0.351	0.885	0.996	0.834	0.781	0.247	0.852	0.912	0.951	0.489	1		
	Pb	-0.052	0.276	0.606	0.515	0.120	0.065	0.635	0.007	0.437	0.700	0.453	0.063	0.074	0.858	0.795	0.647	0.498	0.944	0.763	0.973	0.796	0.485	0.942	0.708	1	
	Zn	-0.696	-0.024	0.808	0.171	0.256	-0.359	0.369	0.257	0.095	0.358	0.126	0.099	-0.226	0.533	0.754	0.939	0.626	0.887	0.469	0.868	0.791	0.832	0.496	0.952	0.752	1

Fig. 7 Pearson's correlation coefficient analysis, with the mean concentrations of the chemical elements determined in the mineral waters from the Water Parks of Águas de Contendas and Lambari

^{228}Ra (r = − 0.927). On the other hand, in the comparison between the two parks, it is verified strong positive correlations (r = 0.903) between ^{226}Ra from Águas de Contendas and ^{210}Po from Lambari, and between ^{210}Pb from Águas de Contendas and ^{228}Ra from Lambari (r = 0.817).

Another Pearson correlation was performed to verify the correspondence among the chemical elements in both Water Parks, Fig. 7. In this analysis, it was observed a strong correlation directly proportional, above 0.7, between K and Na elements, alkali-metals, in both parks; between Ba and Mg elements, alkaline-earth metals, also in both parks; Ba and Ca and Ca and Mg in Water Park of Lambari, and among transition elements, such as Cr and Ni and Mn and Ni, in both parks.

Conclusions

The Water Park of Lambari presented the highest activity concentrations for ^{226}Ra, ^{228}Ra and ^{210}Pb, while for ^{210}Po the highest concentrations were determined at Water Park Águas de Contendas; and these activity concentrations varied among the seasons without a standard concentration behavior; only ^{228}Ra activity concentrations exceeded the MVA of 0.1 Bq L^{-1} in some samplings. The radionuclides' analyzes in drinking water collected in the cities near the Water Parks prove the natural radioactivity present in the mineral waters studied and the influence of the local geology in the activity concentration, when natural radionuclides are present with values above the background level. The major elements, Na and Fe in Águas de Contendas and Ca and Na in Lambari, presented the highest concentrations and the mineral waters from both Water Parks were classified as soft in relation to THW, presenting values below the MPV for drinking water. The present work presented the first activity concentration results of ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po in the Water Park of Águas de Contendas, as well as concentrations for the elements As, Cd, Cr and Ni. Pearson's correlation coefficient analysis showed few positive correlations among the radionuclides and chemical elements studied, indicating that several factors, such as, study area lithology, the structure of the rocks and the water flow system, among others, may influence the geochemistry behavior of the natural radionuclides and chemical elements in groundwater.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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