BJRS

BRAZILIAN JOURNAL
OF
RADIATION SCIENCES
10-03B (2022) 01-15

Assessment of natural radioactivity in bottled mineral water from Brazil

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ABSTRACT

The approach taken in the WHO Guidelines for controlling radiological hazards in public water supplies has two stages. The first is an initial screening for gross alpha and beta activity to determine whether the activity concentrations are below levels at which no further action is required; and if these screening levels are exceeded, investigation of the concentration of individual radionuclides and comparison with specific guidance levels. Mineral water is obtained directly from natural sources or by extracting groundwater, it is characterized by the quantity of mineral salts, trace elements and other constituents. The bottled water industry is present in the major regions of the world, and the population consumption has increased every year in Brazil and world, also increasing new mineral water mining sites, so it is necessary to constantly check the amount of radioactivity in mineral water. The aim of this study is to quantify the concentration of the natural radionuclides in the bottled mineral water consumed in São Paulo city. The bottled mineral water samples were purchased in a supermarket in São Paulo, with 23 different brands. The determination of gross alpha and beta activity concentration in bottled mineral water by Liquid Scintillation Counting measurement was performed using a 1220 Quantulus™ Ultra Low-Level Liquid Scintillation Spectrometer. The natural radionuclides (^{226}Ra, ^{228}Ra, ^{210}Pb and ^{40}K) were measured by gamma spectrometry, using an HPGe detector. The results obtained in this study can be used for a database on bottled mineral water radioactivity from Brazil.

Keywords: **mineral water, natural radionuclides, Liquid Scintillation Counting, gamma spectrometry.**

ISSN: 2319-0612
DOI: https://doi.org/10.15392/2319-0612.2022.1795
Submitted: 2022-01-28
Accepted: 2022-09-21

1. INTRODUCTION

Humans are exposed to natural radiation; source of external and internal exposure of radiation which includes cosmic rays and natural radionuclides in soil, air, food and drinking water. The exposures of radiation are different in each region of world, external exposure from the soil and cosmic rays is associated with gamma radiation and internal exposure with air, food and drinking water [1]. UNSCEAR [2] estimated that exposure to natural sources contributes more than 70% of the radiation dose and the world mean from natural sources is 2.4 $mSv.y^{-1}$.

In Brazil, the Guideline for controlling radioactivity levels in drinking water is established by MS and CNEN [3, 4], according to this Guidelines provide the recommendations of the World Health Organization (WHO) [5]. The Guidelines provide the recommendations for managing the risk from hazards in public water supplies about the safety of drinking water in several aspect; the chapter 9 discusses specifically practices on radiological aspects.

The screening levels for drinking water below which no further action is required: 0.5 Bq L^{-1} for gross alpha activity and 1 Bq L^{-1} for gross beta activity, with these screening values the individual dose is 0.1 mSv y^{-1}. The screening level if exceeded for gross alpha or gross beta activity is necessary additional investigation. When the gross beta screening level is exceeded, the ^{40}K contribution must be subtracted. The concentrations activity after ^{40}K subtraction remain above screening levels, another sample should be collected and analyzed, if the values continue above the screening level, investigation of individual radionuclide concentrations will be required [3, 4, 5].

The drinking water can contain radionuclides of natural origin, including the ^{40}K, and natural series of ^{232}Th and uranium (^{238}U and ^{235}U), in particular ^{226}Ra, ^{228}Ra, ^{210}Pb, ^{238}U and ^{234}U [5]. The geological and geochemical factors that influence the concentration of radionuclides in drinking water [6].

The classical approach gross alpha and gross beta consists of evaporating a known volume of water, measure the activity concentration of the residue in a glass flow proportional counter or in a liquid scintillation counting (LSC) coupled to alpha/beta discrimination, which allows a rapid and simple determination of gross alpha and beta activities, which are simultaneously measured [7, 8].

LSC is a technique in which the sample is mixed to the liquid scintillation, forming a scintillation solution, capable of converting the kinetic energy of nuclear emissions into light photons. The interaction of the emissions occurs in the liquid solution, producing excitement with emission of photons of ultraviolet radiation [7].

The gamma ray spectrometry is a technique used for determining the of gamma-ray emitter radionuclides, resulting in a count vs. energy spectrum with good resolution, being able to discriminate and quantify the radionuclide in the spectrum [8].

This paper presents activity concentrations gross alpha, gross beta, natural radionuclides (^{226}Ra, ^{228}Ra, ^{210}Pb and ^{40}K) and assessment of effective dose determined in 23 different brands of bottled mineral waters from Brazil (Minas Gerais, Paraná, Rio de Janeiro, Rio Grande do Sul and São Paulo States), the bottled mineral waters were purchased in São Paulo city supermarkets. The determination of gross alpha and beta by liquid scintillation counting (LSC) and ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{40}K were measured by gamma spectrometry, using an HPGe detector.

The world consumption of bottled water in 2017 was 377 billion liters, 8.1% higher than in 2016. The average rate of global growth in the period 2012-2017 was 6.4%, with Brazil presented the annual mean rate of 4.7% [9].

Godoy et al. [10] analyzed ^{226}Ra, ^{228}Ra and ^{210}Pb in bottled mineral of each Brazilian geographical region (Southeast, Northeast, South, Middle-west and North), with a range < 0.002 to 0.22 Bq L^{-1} for ^{226}Ra, < 0.009 to 1.02 Bq L^{-1} for ^{228}Ra and range of 0.011 to 0.51 Bq L^{-1} for ^{210}Pb.

Oliveira et al. [11] determined activity concentration bottled mineral waters commercially available in Brazil (São Paulo and Minas Gerais States), with a range < 2.2 to 647 mBq L^{-1} for ^{226}Ra, 12 to 741 mBq L^{-1} for ^{228}Ra and for ^{210}Pb with a range of < 4.9 to 85 mBq L^{-1}.

Filho et al. [12] determined gross alpha and beta in commercial mineral water from springs of Beberibe aquifer of Metropolitan Region of Recife, Pernambuco State, Brazil, with a range < 20 to 85 mBq L^{-1} for gross alpha, < 60 to 330 mBq L^{-1} for gross beta.

The database for drinking water (Table 15) of UNSCEAR [1] present activity concentration values, with a range were 0.2 to 49000 mBq L^{-1} for ^{226}Ra; 0.5 to 570 mBq L^{-1} for ^{228}Ra and 0.1 to 21000 mBq L^{-1} for ^{210}Pb.

2. MATERIALS AND METHODS

The ultra-low level liquid scintillation counting (LSC) coupled to alpha–beta discrimination was chosen for determine the gross alpha and beta activity concentration [13]. This method allows rapid and simple determination of gross alpha and beta activities, which simultaneously measured through alpha/beta discrimination technique. The initial step of pre-concentration of the mineral water, consist of concentrate 2 L of water on a hot plate, at a controlled temperature of 80°C, to a final volume of 50 mL, an aliquot of 5 mL of the same final solution is mixed with 15 mL of the scintillation solution (Optiphase Hisafe 3) in the polyethylene vial. The sample were counted on a liquid scintillation counter for 240 minutes. The vials used must be counted with ultrapure water, before the analysis for the evaluation of the background, which implies in an increase of two hours in the total time necessary to perform the analysis. As recommended by ISO 11704 [14], the 20 mL vials used for the measurements were made of low diffusion polyethylene (HDPE), since it presents the best cost benefit over others like polyethylene coated with Teflon (PTFE), glass and low background glass [15].

The equipment used for the measurement of gross alpha and beta activities were 1220 Quantulus™ Ultra Low-level Liquid Scintillation Spectrometer coupled to alpha–beta discrimination [16]. For alpha counting, the counting window was defined in channel 400 to 900, considering alphas with a wide energy range from 3 MeV to 7 MeV, which include most probable natural emitting radionuclides. The counting window for beta was defined in channel from 400 to 1000, where only beta emitters with energy higher than 0.2 MeV are considered natural radionuclides ^{228}Ra and ^{40}K [14]. The background radiation was estimated by measuring ultrapure water.

The mineral water sample remaining was packed 100 mL polyethylene bottle for gamma spectrometry analysis; completed the volume of the counting geometry, sealed, and stored for 30 days to enable the radioactive equilibrium between ^{226}Ra and its decay products.

The measurements of activity concentration of ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{40}K in the bottled mineral waters were of gamma spectrometry analysis, using an HPGe detector GX2518 detector of 25% relative efficiency for the peak of 1332 keV of the ^{60}Co, with associated electronics and coupled to a microcomputer. For the determination of ^{226}Ra it is assumed that it is in equilibrium with ^{214}Pb

and ^{214}Bi. Its activity is determined by the line of its ^{214}Pb decay products, which emits gamma energies of 295.2 keV and 351.9 keV, and ^{214}Bi, which emits gamma energies of 609.3 keV and 1120.3 keV, respectively. ^{228}Ra was determined by measuring peak intensity of 911.07 keV and 969.11 keV for ^{228}Ac. ^{210}Pb was determined directly through its 46.5 keV line. The content of 40K was determined by measuring its 1460 keV gamma rays. The gamma spectra were obtained by the multichannel emulator program Maestro [17] and were analyzed with the WinnerGamma program on the InterWinner platform [18]. The counting time was determined using the model proposed by Nisti [19]. The background radiation was estimated by measuring ultrapure water. The detector was calibrated using source with radionuclides certified by Amersham International. The intensities of gamma transitions have been obtained in the literature [20].

The determination of the minimum detectable concentration (MDC) for both methods was using the model proposed by Currie [21]. The performance of the gamma spectrometry and LSC methods was evaluated by participating in Proficiency Tests (PT) organized by Instituto de Radioproteção e Dosimetria IRD/CNEN [22], indicating good performance of laboratory for the methods used in the paper. The annual effective dose was calculated using the following equation 1 [23]:

$$E = (C_i \times Q \times DC) \quad (1)$$

Where:

E is annual effective dose (Sv year ^{1}),
C_i is the activity concentrations of radionuclide i (Bq L^{-1}),
Q annual ingested volume of drinking-water, assumed to be 730 liters $year^{-1}$ (equivalent to the standard World Health Organization drinking water consumption rate of 2 liters day^{-1}),
DC is effective dose coefficients (Sv Bq^{-1}).

The effective dose coefficients (Sv Bq^{-1}) for adult are presented in Table 1 [23].

Table 1. Effective dose coefficients ^{210}Pb, ^{226}Ra and ^{228}Ra (Sv Bq^{-1}) for adult

	^{210}Pb	^{226}Ra	^{228}Ra
Sv Bq^{-1}	6.9 10^{-7}	2.8 10^{-7}	6.9 10^{-7}

The chemical composition (mg L^{-1}) reported on the bottled mineral water packaging are presented in Table 2 and 3.

Table 2. Chemical composition (mg L^{-1}) reported on the bottled mineral water packaging

Sample	pH (25°C)	Maches	Barium	Bicarbonate	Calcium	Chloride	Strontium
1	7.07	17.77	0.018	104.87	17.4	1.03	0.159
2	6.70	9.18	0.098	34.55	17.3	9.26	n.i.
3	6.38	n.i.	0.022	77.71	13.6	2.31	0.203
4	6.24	11.37	n.i.	29.95	5.73	0.86	0.121
5	7.10	13.50	0.048	126.84	26.5	2.23	0.769
6	6.69	n.i.	0.071	45.83	9.84	3.32	0.139
7	7.37	n.i.	0.133	106.53	3.714	1.40	0.041
8	7.10	13.50	0.048	126.84	26.5	2.23	0.769
9	7.00	n.i.	0.051	27.65	6.36	n.i.	0.023
10	7.91	7.71	0.028	108.51	17.6	n.i.	0.014
11	6.13	18.82	0.032	12.56	2.304	0.40	0.049
12	6.70	9.18	0.098	94.55	17.3	9.26	n.i.
13	8.10	9.16	0.030	105.78	17.3	0.18	0.018
14	7.15	n.i.	0.034	159.93	37.2	0.82	0.250
15	7.60	n.i.	n.i.	146.62	24.88	0.64	0.019
16	5.93	11.23	n.i.	25.27	1.483	0.33	0.156
17	6.42	8.20	0.060	68.00	11.7	0.61	0.141
18	7.27	n.i.	0.062	169.14	31.0	10.85	0.025
19	7.10	13.50	0.048	126.84	26.5	2.23	0.769
20	5.25	n.i.	0.408	286.73	28.169	1.61	0.053
21	6.15	5.63	0.072	45.18	6.62	1.87	0.074
22	10.0	n.i.	n.i.	78.43	1.25	0.2	0.037
23	7.1	13.50	0.048	126.84	26.5	2.23	0.769

ISSN: 2319-0612
DOI: https://doi.org/10.15392/2319-0612.2022.1795
Submitted: 2022-01-28
Accepted: 2022-09-21

n.i.: not informed

Table 3. Chemical composition (mg L^{-1}) reported on the bottled mineral water packaging

Sample	Fluoride	Phosphate	Magnesium	Nitrate	Potassium	Sodium	Sulfate
1	0.13	0.11	2.78	0.08	3.82	16.2	6.98
2	0.06	n.i.	7.21	10.43	2.9	11.5	2.39
3	0.07	0.27	2.69	2.88	3.78	9.51	1.1
4	0.09	0.25	0.896	5.58	2.39	4.64	0.53
5	1.02	0.12	1.37	0.54	2.71	24.6	18.53
6	0.12	0.23	1.714	10.29	2.783	7.345	0.25
7	0.93	0.86	1.205	n.i.	2.765	35.105	1.69
8	1.02	0.12	1.37	0.54	2.71	24.6	18.53
9	0.03	0.39	0.731	1.31	3.91	0.412	0.6
10	0.07	n.i.	10.1	0.24	1.36	1.07	n.i.
11	0.07	n.i.	0.356	0.01	1.102	1.991	0.40
12	0.06	n.i.	7.21	10.43	2.9	11.5	2.39
13	0.04	n.i.	9.78	0.74	1.44	1.16	0.12
14	0.12	0.06	7.99	0.51	2.94	6.46	10.64
15	0.02	n.i.	14.29	n.i.	0.542	1.079	n.i.
16	0.09	n.i.	0.33	0.25	2.243	6.351	0.31
17	0.27	n.i.	5.39	1.30	3.52	2.9	3.88
18	0.11	n.i.	16.5	5.15	3.99	4.18	1.11
19	1.02	0.12	1.37	0.54	2.71	24.6	18.53
20	0.15	n.i.	12.491	1.50	31.346	36.04	2.17

21	0.03	0.16	3.31	14.18	2.97	8.01	0.18
22	0.33	n.i.	0.09	0.5	0.21	75.81	0.2
23	1.02	0.12	1.37	0.54	2.71	24.6	18.53

n.i.: not informed

3. RESULTS AND DISCUSSION

The counting efficiency for gross alpha and beta by LSC was determined using standard solutions of ^{241}Am and ^{90}Sr+^{90}Y, and background radiation was estimated by measuring ultrapure water in vial.

The PSA parameter value that minimizes total interference was determined for the scintillating solution and proportion 5:15 (sample and cocktail) studied. The alpha interference was defined as the alpha fraction of alpha particles observed in the beta spectrum when measuring an alpha pure emitter; and beta interference is the fraction of beta particles observed in the alpha spectrum when measuring a beta pure emitter, and total interference is the sum alpha and beta. The results obtained for the PSA optimization in the windows chosen are presented in Fig. 1.

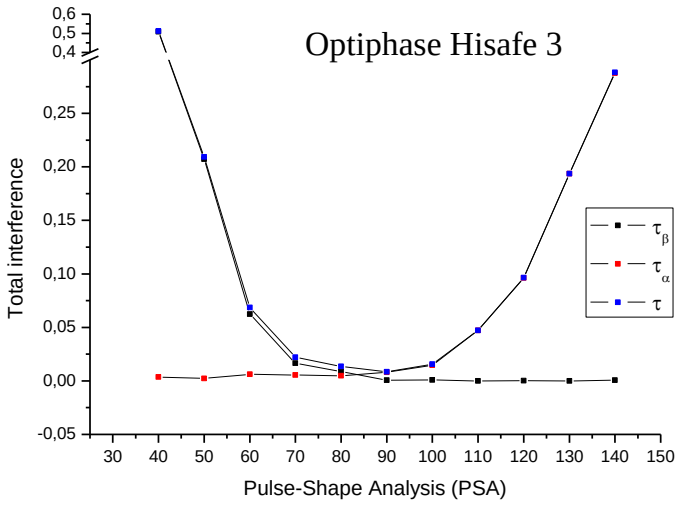

Fig. 1 Alpha, beta and total interference for the Optiphase Hisafe 3 cocktail in proportion 5:15

The gross alpha and beta of bottled mineral waters were counted in duplicate. The activity concentrations gross alpha and beta obtained in the bottled mineral waters are presented in the Table 3.

ISSN: 2319-0612
DOI: https://doi.org/10.15392/2319-0612.2022.1795
Submitted: 2022-01-28
Accepted: 2022-09-21

Table 3. Concentration of gross alpha and beta in the bottled mineral waters (Bq L^{-1})

Sample	State of Origin	Alpha (A)	Alpha (B)	Mean Alpha	Beta (A)	Beta (B)	Mean Beta
1	SÃO PAULO	0.095	0.091	0.093±0.003	0.20	0.20	0.20±0.01
2	SÃO PAULO	0.023	0.02	0.022±0.002	0.21	0.21	0.21±0.01
3	SÃO PAULO	0.02	0.024	0.022±0.003	0.13	0.12	0.12±0.01
4	MINAS GERAIS	0.016	0.015	0.016±0.001	0.095	0.093	0.094±0.001
5	SÃO PAULO	0.04	0.031	0.036±0.006	0.15	0.12	0.13±0.02
6	SÃO PAULO	< 0.01	< 0.01	< 0.01	0.095	0.11	0.10±0.01
7	SÃO PAULO	0.016	< 0.01	0.016	0.10	0.10	0.10±0.01
8	SÃO PAULO	0.038	0.036	0.037±0.001	0.12	0.10	0.11±0.01
9	SÃO PAULO	< 0.01	< 0.01	< 0.01	0.20	0.20	0.20±0.01
10	SÃO PAULO	0.035	0.043	0.039±0.006	0.096	0.094	0.095±0.001
11	RIO DE JANEIRO	0.018	0.019	0.019±0.001	< 0.05	< 0.05	< 0.05
12	SÃO PAULO	0.027	0.02	0.024±0.005	0.12	0.13	0.13±0.01
13	SÃO PAULO	0.031	0.036	0.034±0.004	0.078	0.084	0.081±0.004
14	SÃO PAULO	0.045	0.042	0.044±0.002	0.15	0.15	0.15±0.01
15	RIO GR. DO SUL	0.012	0.01	0.011±0.001	< 0.05	< 0.05	< 0.05
16	SÃO PAULO	0.019	0.018	0.019±0.001	0.21	0.20	0.20±0.01
17	SÃO PAULO	**0.104**	**0.116**	**0.11±0.01**	0.21	0.20	0.20±0.01
18	SÃO PAULO	0.027	0.016	0.022±0.008	0.14	0.13	0.14±0.01
19	SÃO PAULO	0.037	0.03	0.034±0.005	0.18	0.13	0.15±0.04
20	MINAS GERAIS	0.037	0.024	0.031±0.009	**1.61**	**1.59**	**1.60±0.01**
21	SÃO PAULO	< 0.01	< 0.01	< 0.01	0.15	0.16	0.16±0.01
22	PARANÁ	0.077	0.079	0.078±0.001	0.22	0.22	0.22±0.01
23	SÃO PAULO	0.023	0.033	0.028±0.007	0.14	0.13	0.14±0.01

The activity concentrations ^{210}Pb, ^{226}Ra, ^{228}Ra and ^{40}K obtained in the bottled mineral waters are presented in the Table 4.

Table 4. Concentration of ^{210}Pb, ^{226}Ra, ^{228}Ra and ^{40}K in the bottled mineral waters (Bq L^{-1})

Sample	**State of origin**	**^{210}Pb**	**^{226}Ra**	**^{228}Ra**	**^{40}K**
1	SÃO PAULO	< 0.31	< 0.08	< 0.13	< 0.52
2	SÃO PAULO	< 0.26	< 0.08	< 0.15	< 0.60
3	SÃO PAULO	< 0.33	< 0.08	< 0.13	< 0.57
4	MINAS GERAIS	< 0.33	< 0.08	< 0.13	< 0.55
5	SÃO PAULO	< 0.28	< 0.08	< 0.13	< 0.58
6	SÃO PAULO	< 0.33	0.07±0.01	0.11±0.03	< 0.54
7	SÃO PAULO	< 0.29	< 0.08	< 0.13	< 0.60
8	SÃO PAULO	< 0.31	< 0.09	< 0.15	< 0.62
9	SÃO PAULO	< 0.36	< 0.08	< 0.14	< 0.60
10	SÃO PAULO	< 0.35	< 0.08	0.13±0.03	< 0.60
11	RIO DE JANEIRO	< 0.27	< 0.08	< 0.16	< 0.65
12	SÃO PAULO	< 0.32	< 0.08	< 0.15	< 0.64
13	SÃO PAULO	< 0.25	< 0.08	< 0.13	< 0.53
14	SÃO PAULO	< 0.25	< 0.07	< 0.12	< 0.54
15	RIO GR. DO SUL	< 0.26	0.07±0.02	0.11±0.03	< 0.54
16	SÃO PAULO	< 0.28	< 0.08	< 0.13	< 0.60
17	SÃO PAULO	< 0.26	**0.32±0.03**	< 0.12	< 0.58
18	SÃO PAULO	< 0.25	0.06±0.01	0.16±0.03	< 0.53
19	SÃO PAULO	< 0.26	< 0.07	< 0.13	< 0.53
20	MINAS GERAIS	< 0.36	0.08±0.01	**0.19±0.03**	**1.32±0.16**
21	SÃO PAULO	< 0.28	< 0.07	0.09±0.02	< 0.57
22	PARANÁ	< 0.29	< 0.08	< 0.13	< 0.60
23	SÃO PAULO	< 0.29	< 0.08	< 0.13	< 0.60

The highest gross alpha and ^{226}Ra activity concentration was mineral water sample 17, but did not exceed the screening level.

The concentrations gross beta in the bottled mineral waters (Table 3), the sample 20 was exceeded screening levels for drinking water; WHO [5] guidelines for controlling radiological hazards in public water supplies becomes investigation necessary of the concentration of individual radionuclides and comparison with specific guidance levels. All other samples were below the screening level for gross beta in drinking water.

The activity concentration obtained in this work, with a range were < 0.07 to 0.32 Bq L^{-1} for ^{226}Ra; < 0.12 to 0.19 Bq L^{-1} for ^{228}Ra and < 0.52 to 1.32 Bq L^{-1} for ^{40}K. For the ^{210}Pb, all results of the activity concentration were below the MDC.

The activity concentration values in this work are in the range of the literature values [1, 10, 11].

The concentrations of activity of sample 20 were 1.32±0.16 Bq L^{-1} for ^{40}K and 0.19±0.03 Bq L^{-1} for ^{228}Ra, the total concentration of the beta emitting radionuclides was of 1.51±0.16 Bq L^{-1}, concentration similar the value of gross beta (1.60±0.01 Bq L^{-1}), WHO [5] recommendation: the contribution of ^{40}K to beta activity should be subtracted. Thus resulting, the value of the concentration of the beta below screening levels for drinking water for sample 20.

Tables 1 and 2 show the chemical composition in bottled mineral water, reported on the packaging; comparing the values of bottled mineral waters studied the sample 20 presented the highest values of barium, potassium and highest activity concentrations of ^{228}Ra and ^{40}K (Table 4), ^{228}Ra has chemical characteristics similar to barium, so the result is consistent.

Sample 20, the reported value of stable potassium was of 31.346 mg L^{-1}; and only sample with activity concentration ^{40}K above the MDC, obtaining the value of 42±5 Bq g^{-1}. WHO [5] defines that the beta activity of ^{40}K is 27.9 Bq g^{-1} in relation to stable potassium, demonstrating that the value found is the same order of magnitude reported by WHO**.**

The annual effective dose obtained in this work, with a range were 1.4 10^{-2} to 6.5 10^{-2} mSv y^{-1} for ^{226}Ra and < 5.5 10^{-2} to 9.6 10^{-2} mSv y^{-1} for ^{228}Ra. ^{210}Pb was not calculated annual effective dose, all results of the activity concentration were below the MDC. The ^{40}K was not calculated because of the homeostatic control of the human body.

4. CONCLUSION

The highest gross alpha concentration was mineral water sample 17, but did not exceed the screening level. Gross beta the sample 20 was exceeded screening levels for drinking water, therefore necessary additional investigation.

The activity concentration obtained in this work, with a range were < 0.07 to 0.32 Bq L^{-1} for ^{226}Ra; < 0.12 to 0.19 Bq L^{-1} for ^{228}Ra and < 0.52 to 1.32 Bq L^{-1} for ^{40}K. For the ^{210}Pb, all results of the activity concentration were below the MDC.

The concentrations of activity of sample 20 were 1.32±0.16 Bq L^{-1} for ^{40}K and 0.19±0.03 Bq L^{-1} for ^{228}Ra, the total concentration of the beta emitting radionuclides was of 1.51 ± 0.16 Bq L^{-1}, concentration similar the value of gross beta (1.60±0.01 Bq L^{-1}), WHO [5] recommendation: the contribution of ^{40}K to beta activity should be subtracted. Thus resulting, the value of the concentration of the beta below screening levels for drinking water for sample 20.

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Finally, the results of this paper showing that the all bottled mineral waters analyzed were below screening levels for drinking water recommended by WHO.

The results in this paper can be used for a database on concentrations gross alpha and beta, natural radionuclides and effective dose in bottled mineral water consumed in the city of São Paulo.

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