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Assessment of Natural Radioactivity in Bottled Mineral Water from Brazil

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1. Introduction

The radiation exposure of naturally occurring radionuclides through drinking water comprises radioactive elements from the ^{238}U and ^{232}Th series, such as ^{226}Ra, ^{228}Ra and ^{210}Pb. In addition to these radionuclides, ^{40}K, another natural radionuclide, is also found in water. The concentration of radionuclides in water is influenced by geological and geochemical factors [1].
In Brazil, the Guideline for controlling radioactivity levels in drinking water was established by Ministério da Saúde (M.S.) [2], in agreement with the recommendations of the World Health Organization (WHO) [3]. The Guideline provides general recommendations for safe drinking water, as well as guideness for managing the risks due to hazards found in public water supplies. The screening levels for drinking water, below which no further action is required, are: 0.5 Bq L^{-1} for gross alpha activity and 1 Bq L^{-1} for gross beta activity, resulting in individual dose of 0.1 mSv y^{-1}.
This paper presents gross alpha and gross beta activity concentrations, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{40}K natural radionuclides activity concentrations and the effective dose in different brands of mineral water regarding their consumption.

2. Methodology

Twenty three different brands of mineral bottled water were purchased in several supermarkets in Sao Paulo city. These samples contemplated mineral waters from different Brazilian states, as Minas Gerais, Paraná, Rio de Janeiro, Rio Grande do Sul and São Paulo. Gross alpha and gross beta activity concentrations were measured using liquid scintillation counting (LSC); the activity concentrations for ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{40}K were measured by gamma spectrometry, using an HPGe detector.
The initial step of pre-concentration of the mineral water, consist of concentrating 2 L of water on a hot plate, at a controlled temperature of 80°C, to a final volume of 50 mL. A 5 mL aliquot from the sample is than mixed with 15 mL of the scintillation solution (Optiphase Hisafe 3) in the polyethylene vial. The water sample is counted on a liquid scintillation counter for 240 minutes. 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 [4].
For alpha counting, the counting window was defined for channels 400 to 900, considering alphas with a wide energy range from 3 MeV to 7 MeV. Beta window was defined for channels 400 to 1000, where only beta emitters with energy higher than 0.2 MeV are considered, as it is for natural radionuclides such as ^{228}Ra and ^{40}K [5]. The background radiation was estimated using ultrapure water measurements.
The mineral water sample that remained was accommodated in a polyethylene bottle, sealed and stored for 30 days to ensure secular radioactive equilibrium between ^{226}Ra and its decay products.
Measurements of activity concentrations from ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{40}K were obtained using gamma spectrometry analysis, with an HPGe detector (GX2518 detector with 25% relative efficiency considering the 1332 keV peak energy from ^{60}Co). The counting time was determined using the model proposed by Nisti et al. [6]. The determination of the minimum detectable concentration (MDC) for both methods was made using the model proposed by Currie [7]. The annual effective dose and effective dose coefficients was calculated using the ICRP [8].

3. Results and Discussion

The gross alpha and beta of mineral bottled waters were counted in duplicate. The activity concentrations gross alpha and beta obtained in the bottled mineral waters are presented in the Table 1.

Table 1. Gross alpha and beta concentrations in the mineral bottled 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 gross beta concentrations for sample 20 exceeded the screening levels for drinking water. The WHO [3] guidelines for controlling radiological hazards in public water supplies states that investigation is than necessary for the concentrations of individual radionuclides and their comparison to recommended levels. All other samples were below the screening level for gross beta in drinking water. Bottled mineral water is not considered public water, but with the increasing consumption and no specific regulation available, considering the radiological risk, M.S. [2] and WHO [3] were used as recommendations in this work.

The activity concentrations for ^{210}Pb, ^{226}Ra, ^{228}Ra and ^{40}K obtained in are presented in the Table 2.

Table 2. Concentrations ^{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 activity concentrations levels obtained ranged from < 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. All activity concentration results were below the MDC for the ^{210}Pb. The activity concentration values in this work agree with the range found in the literature [9,10,11].

The activity concentrations from 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 beta emitting radionuclides was of 1.51 ± 0.16 Bq L^{-1}; similar to the concentration value of gross beta in Table 1 (1.60 ± 0.01 Bq L^{-1}). The WHO [3] recommendation: the contribution of ^{40}K to beta activity should be subtracted, thus resulting in the value below screening levels for drinking water for bottled mineral water.

The annual effective dose obtained in this work, ranged from 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. Effective dose from ^{210}Pb was not calculated, since all results were below the MDC. Effective dose from ^{40}K was not calculated once potassium participates in the homeostatic control of the human body.

4. Conclusions

The activity concentration obtained in this work, ranged from < 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. Activity concentrations for ^{210}Pb were found below the MDC. The activity concentrations from 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 beta emitting radionuclides was of 1.51 ± 0.16 Bq L^{-1}, similar to the value of gross beta

(1.60 ± 0.01 Bq L^{-1}). According to the WHO (2011) recommendation: the contribution of ^{40}K to beta activity should be subtracted, thus resulting in the value below screening levels for drinking water for bottled mineral water.

The annual effective dose obtained in this work, ranged from 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. Effective dose from ^{210}Pb was not calculated, since all results were below the MDC. Effective dose from ^{40}K was not calculated once potassium participates in the homeostatic control of the human body.

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

To ensure the quality of the bottled mineral water consumed by the population, continuous monitoring is recommended to check the a screening levels, specially considering the constant increase in its consumption and new sites of production.

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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