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URANIUM, THORIUM, GROSS ALPHA AND GROSS BETA ASSESSMENT IN FOUNTAIN WATERS IN TOWNS OF THE IRON QUADRANGLE, BRAZIL

Cláudia A. Ferreira[1*], Helena E. L. Palmieri[1], Maria Ângela de B. C. Menezes[1], Renata D. A. Chaves[1] and Ilza Dalmázio[1]

[1]Centro de Desenvolvimento da Tecnologia Nuclear/Comissão Nacional de Energia Nuclear, CDTN/CNEN
[*]Pós-graduação em Ciência e Tecnologia das Radiações, Minerais e Materiais
Av. Presidente Antônio Carlos, 6627 Campus UFMG, CEP 31.270-970
Caixa Postal 941, CEP 30161-970, Belo Horizonte, MG
cferreiraquimica@yahoo.com.br, help@cdtn.br, menezes@cdtn.br, rda@cdtn.br, id@cdtn.br

ABSTRACT

The Iron Quadrangle region is known worldwide for its diversity, both ores and rock types, which record a long and important period of Earth's history. For thousands of years erosive processes have exposed ancient rocks, Archean and Proterozoic, in this region. The concentration of uranium, thorium, gross alpha and gross beta activities has been assessed in 34 fountains water samples collected from different towns in the Iron Quadrangle. The results obtained were compared to values established by CONAMA nº 396/2008 and Decreet nº 2914/2011 by the Ministry of Health. For Th in water consumption there is no value established in the Brazilian legislation and the concentrations in all samples were lower than 0.01 µg L^{-1}. For uranium, the values ranged from less than 0.002 to 0.61 µg L^{-1}, and all results were lower than the value allowed of 15 µg L^{-1} and 30 µg L^{-1} established by the legislations above, respectively. The results for the radiation levels of gross alpha and gross beta activity in some fountains waters were slightly above the limits (0.5 Bq L^{-1} and 1.0 Bq L^{-1}) established by CONAMA nº 396/2008 and Decreet nº 2914/2011, respectively.

1. INTRODUCTION

Water essential to human health and used for consumption by the population must meet potability standards. In Brazil, Decreet n° 2914 of December 12, 2011 by the Ministry of Health established the maximum standard for radioactivity in drinking water of 0.5 Bq L^{-1} for gross alpha and 1.0 Bq L^{-1} for gross beta. According to the Decreet specific analysis should be performed if gross alpha and gross beta limits are exceeded due to possible presence of radionuclides [1,2]. For uranium the new Decreet by the Ministry of Health established the maximum concentration of 30 µg L^{-1} [2]. For Th in water consumption, there is no value established in Brazilian legislations. Therefore, a work needs to be done to propose a database including the various levels found.

At the National Environment Council (CONAMA), number 396 of April 3 2008, maximum levels of potentially harmful substances are established. For uranium in water consumption, the maximum allowed value is 15 µg L^{-1}. It is understood that all waters which naturally or artificially occurring in underground is groundwater [3].

The Iron Quadrangle region is known worldwide for its diversity, both ores and rock types, which record a long and important period of Earth's history. For thousands of years erosive processes have exposed ancient rocks, Archean and Proterozoic, in this region occupying the

southeast portion of the São Francisco Craton, the Iron Quadrangle encompasses four main lithostratigraphic units: the granite-gneissic metamorphic complexes; the Archean greenstone belt sequence, known as the Rio das Velhas Supergroup; the Paleoproterozoic metasediments and metavulcanics of the Minas Supergroup [4]; and the metasedimentary rocks of the Itacolomi Group. The main gold deposits are found along shear zones which cut the basic volcanic rocks and banded iron formations of the Nova Lima Group, base of the Rio das Velhas Supergroup. Gold deposits from Nova Lima and Santa Barbara and Raposos, are mainly associated with pyrite, pyrrhotite and arsenopyrite-bearing banded iron formations. Another situation is exemplified in the Ouro Preto- Mariana district; the Passagem de Mariana deposit is located close to the contact between the rocks of the Nova Lima Group and the Minas Supergroup. This ore deposit contains major amounts of arsenopyrite, either in quartz and carbonate veins or disseminated in banded iron formation, tourmalinite, phyllite and quartzite [4,5].

Due to the available water resources in the Iron Quadrangle region, there are many fountains (Figure 1) used for consumption by the population. However, these water sources are not monitored and the water quality is not evaluated in order to know whether the values meet the Brazilian Legislation. In the past these fountains played an essential and strategic role in supplying potable water to several towns located in the Iron Quadrangle (QF). Until today this water is used by both the local population and tourists who believe in its quality.

The objective of this study is to assess the concentrations of uranium, thorium, gross alpha and gross beta activities in 34 fountain water samples, for human consumption, collected in 18 towns of the Iron Quadrangle, Brazil.

Figure 1: Several fountain waters found in towns located in the Iron Quadrangle (QF).

2. MATERIALS AND METHODS

Fountain water samples were collected in towns spread in the Iron Quadrangle region, Minas Gerais State, Brazil, in August 2011 and March 2012, as described in Table 1. The number in brackets corresponds to each fountain.

For the assessment of gross alpha and gross beta activities, about one liter of water samples was collected and stored in low density polyethylene bottles, previously decontaminated in acidic solution. After collection the samples were immediately acidified with nitric acid by adding 10 mL of 65% (w/w) HNO_3. The natural radioactivity levels (gross α and gross β) were determined by a procedure involving evaporation/Proportional Counter with Gas Flow, GFPC, in the Low Activity Radiochemical Laboratory at Division for Reactor and Analytical Techniques (SERTA), CDTN/CNEN.

Uranium and thorium concentrations were analyzed using inductively coupled plasma mass spectrometry (ICP-MS) according to U.S. EPA method [6]. After collection, the samples were filtered through Millipore 0.45 µm filters and immediately acidified with nitric acid (Fluka) and then stored in acid pre-cleaned, low density polyethylene bottles.

All measurements were carried out using a Perkin Elmer ELAN DRC-e ICP-MS spectrometer (Figure 2) equipped with auto sampler (AS-93plus), sea spray nebulizer, cyclonic spray chamber and quadrupole analyzer and data were obtained using a Dell computer with a Perkin Elmer SCIEX quantitative ELAN Version 3.4 software. Argon gas 99.999% purity, supplied by White Martins, was used for plasma, sample nebulisation, and as auxiliary gas. The concentrations of these elements were analyzed in Laboratory of Mass Spectrometry, SERTA.

Figure 2: ICP-MS ELAN DRC-e (Perkin Elmer) System with Auto Sampler.

Table 1: Towns of the Iron Quadrangle and their UTM coordinates for 34 samples collected in 18 towns of the Iron Quadrangle region

Towns	Fountains	UTM Coordinates		Towns	Fountains	UTM Coordinates	
Santa Luzia	Fonte Camelos (1)	619977	7814150	Ouro Preto	Pilar (18)	655742	7745036
Sabará	Kaquende (2)	624273	7799916	Nova Lima	Bica Mirante da Mata (19)	616897	7788318
	Bica (3)	625938	7800183		Bica na Praça dos Cristais (20)	620125	7789093
Caeté	Bica Serra Piedade (4)	638975	7808555		Vila Lacerda (21)	620352	7788847
	Bica 2 (5)	637982	7811193		Chafariz BNH (22)	619641	7788990
Barão de Cocais	Fonte Três Bicas (6)	657984	7793729		Chafariz do Leão (23)	619169	7787910
	Bica 2 (7)	658185	7795069	Rio Acima	Bica José Paulino (24)	629206	7773733
	Bica 3 (8)	658376	7795557	Raposos	Água Limpa (25)	624207	7791750
Santa Bárbara	Bica Linha de Trem (9)	666395	7790694		Bica Recanto Feliz (26)	625570	7791404
	Parque Recanto Verde (10)	666707	7791686	Mário Campos	Bica Bela Vista (27)	588496	7780158
Catas Altas	Nossa Senhora da Conceição (11)	666402	7779405	Moeda	Bica Gruta (28)	608943	7757430
Mariana	Praça Sé (12)	665308	7745915	Congonhas	Fonte do Moinho (29)	618100	7732018
	Praça Jardins (13)	665196	7745799		Biquinha da Basílica (30)	618884	7731790
Passagem Mariana	Fonte da Glória (14)	662212	7744185	Itabirito	Fonte da Açucena (31)	624547	7759961
Ouro Preto	Bica Lajes (15)	656754	7745299	Lavras Novas	Bica das Rosas (32)	654282	7734941
	D. Pedro II (16)	656083	7745248		Fonte Grande (33)	654742	7735234
	Coimbra (17)	656231	7745089		Fonte da Biquinha (34)	654553	7735545

3. RESULTS AND DISCUSSION

The analytical results of the fountain water samples collected are given in Table 2. For uranium, the values ranged from less than 0.002 to 0.61 µg L^{-1} (August) and less than 0.002 to 0.54 µg L^{-1} (March), but all results were lower than the maximum of 15.0 µg L^{-1} and 30.0 µg L^{-1} allowed by CONAMA nº 396/2008 and Decreet nº 2914/2011, Ministry of Health, respectively. The graph in Figure 3 shows the variation of these concentrations for each collected point.

Table 2: Values of pH, gross alpha, gross beta activities, uranium and thorium concentration in 34 samples collected in 18 towns of the Iron Quadrangle region

August samples						March samples					
Parameters						Parameters					
Fountains	pH	Gross alpha (Bq L^{-1})	Gross beta (Bq L^{-1})	U (µg L^{-1})	Th (µg L^{-1})	Fountains	pH	Gross alpha (Bq L^{-1})	Gross beta (Bq L^{-1})	U (µg L^{-1})	Th (µg L^{-1})
1	5.8	0.15 ± 0.05	0.24 ± 0.04	0.04	<0.01	1	6.0	0.07 ± 0.04	0.30 ± 0.04	0.06	<0.01
2	5.7	0.36 ± 0.05	0.32 ± 0.04	0.02	<0.01	2	5.8	0.26 ± 0.04	0.31 ± 0.04	0.02	<0.01
3	6.1	0.09 ± 0.03	0.09 ± 0.03	<0.002	<0.01	3	6.1	0.06 ± 0.03	0.09 ± 0.03	0.002	<0.01
4	6.2	0.12 ± 0.03	0.14 ± 0.03	0.002	<0.01	4	6.3	0.02 ± 0.02	0.03 ± 0.03	<0.002	<0.01
5	5.0	0.22 ± 0.03	0.26 ± 0.04	0.14	<0.01	5	5.0	0.32 ± 0.04	0.26 ± 0.04	0.16	<0.01
6	5.3	< LD	0.09 ± 0.03	0.002	<0.01	6	5.2	0.07± 0.03	0.08 ± 0.03	0.002	<0.01
7	4.5	0.35 ± 0.04	0.63 ± 0.05	0.10	<0.01	7	4.3	0.29 ± 0.04	0.55 ± 0.04	0.11	<0.01
8	7.7	0.14 ± 0.04	0.16 ± 0.03	0.04	<0.01	8	7.5	0.10 ± 0.03	0.08 ± 0.03	0.04	<0.01
9	5.7	0.44 ± 0.05	0.42 ± 0.04	0.61	<0.01	9	5.8	0.40 ± 0.05	0.47 ± 0.04	0.54	<0.01
10	6.5	0.06 ± 0.03	0.17 ± 0.04	0.26	<0.01	10	6.4	0.07 ± 0.03	0.14 ± 0.03	0.28	<0.01
11	7.2	0.05 ± 0.02	0.05 ± 0.03	<0.002	<0.01	11	5.3	0.03 ± 0.02	0.10 ± 0.03	0.01	<0.01
12	7.4	0.06 ± 0.03	0.11 ± 0.03	0.08	<0.01	12	7.3	0.03 ± 0.02	0.08 ± 0.03	0.04	<0.01
13	7.1	0.07 ± 0.03	0.08 ± 0.03	0.09	<0.01	13	7.0	0.04 ± 0.02	< LD	0.03	<0.01
14	4.8	0.17 ± 0.03	0.20 ± 0.04	0.01	<0.01	14	4.8	0.01 ± 0.02	0.11 ± 0.03	0.01	<0.01
15	6.0	0.19 ± 0.03	0.26 ± 0.04	0.01	<0.01	15	5.7	0.02 ± 0.02	0.04 ± 0.03	0.01	<0.01
16	6.5	0.14 ± 0.03	0.15 ± 0.03	0.01	<0.01	16	4.8	0.10 ± 0.03	0.07 ± 0.03	0.01	<0.01
17	6.4	0.18 ± 0.04	0.23 ± 0.04	<0.002	<0.01	17	6.0	0.03 ± 0.02	0.04 ± 0.03	<0.002	<0.01
18	6.1	0.04 ± 0.02	0.06 ± 0.03	0.02	<0.01	18	6.1	< LD	0.05 ± 0.03	0.02	<0.01
19	6.3	0.10 ± 0.03	0.15 ± 0.03	<0.002	<0.01	19	6.2	< LD	0.04 ± 0.03	<0.002	<0.01
20	6.9	0.17 ± 0.05	0.11 ± 0.03	0.02	<0.01	20	6.8	0.06± 0.02	0.08 ± 0.03	0.02	<0.01
21	6.9	0.21 ± 0.06	0.15 ± 0.04	0.04	<0.01	21	6.8	0.04 ± 0.02	0.07 ± 0.03	0.04	<0.01
22	6.0	0.07 ± 0.02	0.12 ± 0.03	<0.002	<0.01	22	5.8	0.03 ± 0.02	0.08 ± 0.03	0.002	<0.01
23	8.7	0.10 ± 0.03	0.11 ± 0.03	0.03	<0.01	23	8.0	0.05 ± 0.03	0.08 ± 0.03	0.02	<0.01
24	7.2	0.06 ± 0.03	0.13 ± 0.03	<0.002	<0.01	24	6.9	0.10 ± 0.03	0.07 ± 0.03	0.002	<0.01
25	6.0	< LD	0.02 ± 0.03	<0.002	<0.01	25	5.8	0.03 ± 0.02	0.05 ± 0.03	0.002	<0.01
26	6.0	0.02 ± 0.02	0.03 ± 0.03	<0.002	<0.01	26	5.9	0.09 ± 0.03	0.06 ± 0.03	0.002	<0.01
27	4.8	0.03 ± 0.02	0.04 ± 0.03	0.02	<0.01	27	5.0	0.02 ± 0.02	0.10± 0.03	0.01	<0.01
28	5.3	0.06 ± 0.02	0.08 ± 0.03	0.01	<0.01	28	5.2	0.12 ± 0.03	0.07 ± 0.03	0.01	<0.01
29	7.1	0.05 ± 0.02	0.11 ± 0.03	<0.002	<0.01	29	6.9	0.04 ± 0.02	0.10 ± 0.03	<0.002	<0.01
30	6.1	0.07 ± 0.04	0.20 ± 0.04	<0.002	<0.01	30	6.0	0.09 ± 0.04	0.16 ± 0.03	0.004	<0.01
31	6.6	0.02 ± 0.02	0.06 ± 0.03	<0.002	<0.01	31	6.3	0.07 ± 0.02	0.08 ± 0.03	0.004	<0.01
32	4.6	0.41 ± 0.04	1.08 ± 0.06	0.04	<0.01	32	4.6	0.22 ± 0.03	0.96 ± 0.05	0.03	<0.01
33	4.8	0.10 ± 0.03	0.24 ± 0.04	0.01	<0.01	33	4.7	0.07 ± 0.02	0.17 ± 0.03	0.01	<0.01
34	5.1	0.71 ± 0.06	1.80 ± 0.07	0.07	<0.01	34	4.8	0.59 ± 0.05	1.82 ± 0.07	0.06	<0.01

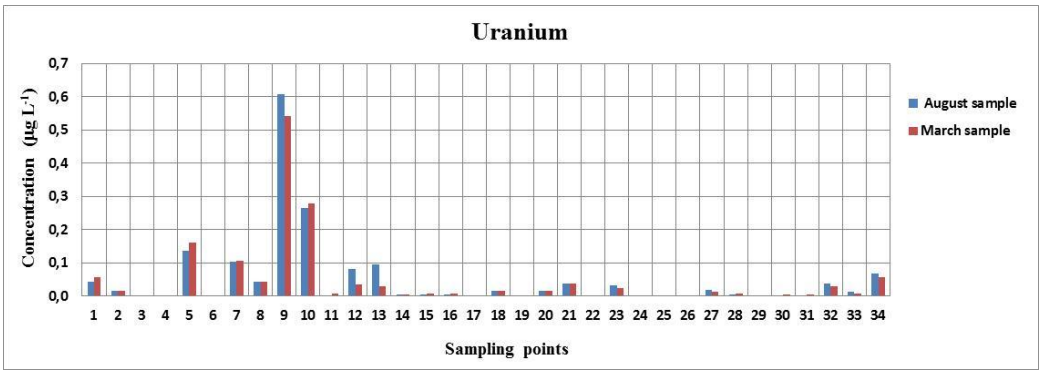

Figure 3: Uranium concentration in fountain water samples in the Iron Quadrangle region.

For Th in water consumption, until now there is no value established in the Brazilian legislation. The concentrations in all samples were less than 0.01 µg L^{-1}, which corresponded to the detection limit of the ICP-MS technique.

The gross alpha values of these waters ranged from 0.02 to 0.71 Bq L^{-1} with a mean of 0.15 Bq L^{-1}. In only one fountain (Biquinha - Lavras Novas, MG) the level of gross alpha activity was above the limit of 0.5 Bq L^{-1} established by the legislation, as seen in Figure 4.

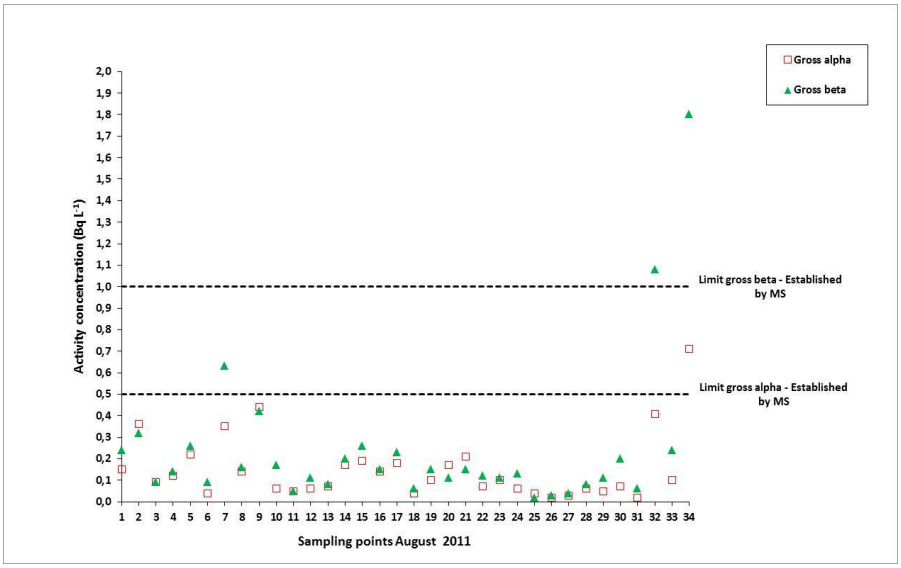

Figure 4: Gross alpha and gross beta activity concentrations (Bq L^{-1}) in fountain waters (August) in towns of the Iron Quadrangle region and limits by Brazilian Health Ministry.

Gross beta activity ranged from 0.02 Bq L^{-1} to 1.80 Bq L^{-1}, with a mean of 0.24 Bq L^{-1}. For gross beta activity, two samples (Bica das Rosas fountain and Biquinha – Lavras Novas, MG) presented radioactivity levels above the 1.0 Bq L^{-1} limits established by the Health Ministry as seen in Figure 5.

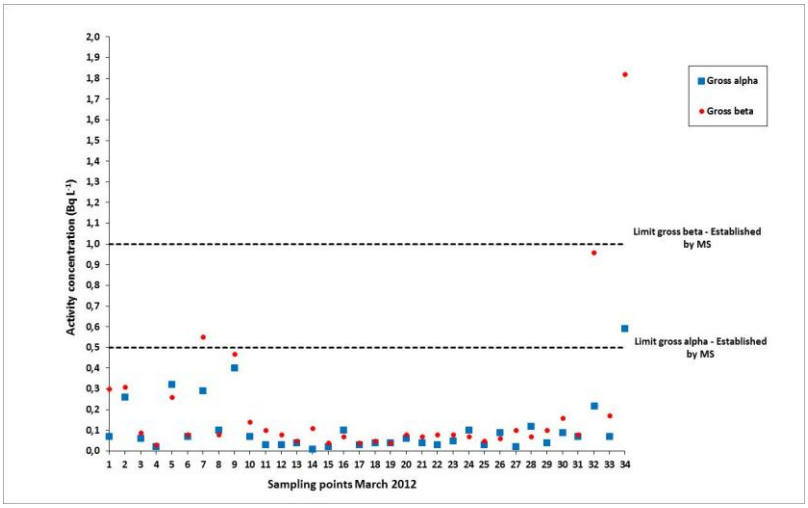

Figure 5: Gross alpha and gross beta activity concentrations (Bq L^{-1}) in fountain waters (March) in towns of the Iron Quadrangle region and limits by Brazilian Health Ministry.

The results of gross alpha values collected in March 2012 ranged from 0.01 e 0.59 Bq L^{-1} (Table 2). As can be seen, in the same sample (Biquinha in Lavras Novas) the limit established is above of the established by CONAMA nº 396/2008 and Decreet nº 2914/2011, Ministry of Health. For gross beta activity the concentrations ranged from 0.03 to 1.82 BqL^{-1}. However, in only Biquinha fountain the limit exceeds the established by the legislations.

4. CONCLUSIONS

The results show that for uranium, the values ranged from less than 0.002 to 0.61 µg L^{-1}, and all results were lower than the value allowed of 15 µg L^{-1} and 30 µg L^{-1} established by the by CONAMA nº 396/2008 and Decreet nº 2914/2011, respectively. For Th in the fountain waters the concentrations in all samples were less than 0.01 µg L^{-1}, which corresponded to the detection limit of the ICP-MS technique.

In only one fountain (Biquinha - Lavras Novas/MG) the level of gross alpha activity was above the limit of 0.5 Bq L^{-1} established by the legislation and for gross beta activity, two samples (Bica das Rosas fountain and Biquinha – Lavras Novas, MG) presented radioactivity levels above the limit of 1.0 Bq L^{-1} established.

The present study pointed out that there is a direct correlation (r~0.90) with gross alpha and gross beta activities in water samples collected in towns spread in the Iron Quadrangle region.

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