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URANIUM CONCENTRATIONS IN THE WATER CONSUMED BY THE RESIDENT POPULATION IN THE VICINITY OF THE LAGOA REAL URANIUM PROVINCE, BAHIA, BRAZIL

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ABSTRACT

The Lagoa Real Uranium Province, situated in South Central Bahia in the region of Caetité and Lagoa Real, is considered the most important monomineralic province of Brazil. The urban population who lives in the proximities of this uranium province in the cities of Caetite, Lagoa Real and Livramento uses public supply water, while the inhabitants of the rural area due to long terms of dry weather use water from wells, cisterns, small dams, reservoirs and dikes which are supplied with the rains. In this work it was determined the concentration of uranium in the water consumed by the rural and urban population living in the proximities of the Lagoa Real Uranium Province. The study comprehends 32 sampling spots spread throughout the region of interest. Samples were collected in January and July 2010, covering superficial, underground and public supply water from the region. The uranium concentrations were determined by an inductively coupled plasma mass spectrometer (ICP-MS). Preliminary results showed that the uranium concentrations in the water from the Lagoa Real Uranium Province varied from 0.064 ± 0.005 $\mu g.L^{-1}$ to $90 \pm 1,5$ $\mu g.L^{-1}$. It was observed that only two of them obtained values higher than the World Health Organization's recommended limit (2011) of 30 $\mu g.L^{-1}$ for maximum uranium concentration in the water for human consumption. For a conclusive evaluation, the uranium concentrations results will be analyzed together with total alpha and beta concentrations determined elsewhere for the same samples.

1. INTRODUCTION

Among the natural radionuclides of terrestrial origin, the ^{40}K and the constituents of the ^{238}U and ^{232}Th series are the greatest source of internal and external exposure in humans. Those natural radionuclides enter the body largely through food and water ingestion [1] being the total exposure per person a result of that ingestion 0.29 mSv, from which 0.17 mSv are due to the ^{40}K and 0.12 mSv are due to the radionuclides of ^{238}U and ^{232}Th series.

Natural uranium is composed by the blend of three isotopes ^{234}U, ^{235}U and ^{238}U in the following proportions: 0.0054%, 0.72% and 99.28%, respectively, and it is found in the whole earth crust in the form of uranium ores, in trace amounts in all kinds of rocks and soils [2, 3], mainly in granitic rocks, metamorphic rocks, monazitic sand, phosphate deposits and minerals such as uranyl and carnotite [4, 5]. In the soil concentrations vary between 1 to 5 $\mu g.g^{-1}$ [6].

The uranium occurs in nature in the valance states +2, +3,+4,+5 and +6, but it comes more frequently in states +4 and +6. In reducing conditions, uranium is present in the form +4 and in such a way it is preferably associated with the solid phase, whereas, in oxidizing conditions U^{6+} it forms the ion uranyl UO_2^{+2}, which is highly soluble. However, in neutral or acid environment the ion uranyl (+6) is susceptible to adsorption by organic matter and oxyhydroxides of Fe. The mobility of the uranium in natural water is associated with the presence of moderate concentrations of complexing ions which avoid the adsorption of the radionuclide therefore facilitating its movement in a watery environment [7].

The uranium transport in oxidant superficial and underground water as a uranyl ion happens in the form of several soluble complexes with dissolved anions. The amounts of uranium in underground water vary a lot depending on litology and/or proximity to ores of that element and they can often be considered anomalous in values over 4 $\mu g.L^{-1.}$ In arid continental conditions there have been registered amounts over several cents of $\mu g.L^{-}$ of U reaching up to 2,000 mg/l or more in the proximities of uranium ores. [6].

Very recent researches showed that the presence of uranium in potable water might make its ingestion into the main access via of the radionuclide to the inside of the human body [8]. In 2004, the World Health Organization recommended a guidance level of 15 $\mu g.L^{-1}$U [9] for the maximum uranium concentration allowed in drinking water. In 2011 [10], the same World Health Organization raised the recommended guidance level to 30 $\mu g.L^{-1}$ U. In Brazil, the National Council of Environment (CONAMA) still recommends 15 $\mu g.L^{-1}$ U as a guidance level for drinking water [11].

In this study, it was determined the uranium concentration in the water consumed by the population resident in the proximities of Lagoa Real Uranium Province. All the water was also analyzed for the evaluation of the total alpha and beta activities concentrations [11]. The importance of this study is due to the fact of drinking water being one of the main entrance pathways of radionuclides in the human body and for Lagoa Real Uranium Province is rich in uranium ore and almost all the hydric resources are used without any analysis or previous radiometric monitoring.

1.1. Characterization of the Field of Study

Brazil is the owner of the seventh greatest geological reserve of uranium in the world, with approximately 310 thousand tons. This reserve is distributed among greater placers, for instance Itatiaia, Ceara (142,500 tons), where the mineral is associated with phosphate and ornamental rocks which are economically exploitable, and Lagoa Real, in Bahia (100,770 tons) and other smaller placers like Gandarela, Minas Gerais, where there is gold associated with the uranium, Rio Cristalino, in Pará; and Figueira, Paraná [12,13].

The Lagoa Real Uranium Province, in the region of Caetite and Lagoa Real, situated in South Central Bahia, constitutes the major uranium ore extraction site currently active in South America and it is considered the most important monomineralic province of Brazil discovered in the 1970's from an aerogamametric survey in the central portion of the Sao Francisco Crato, in the region of the Northern *Espinhaço* [14].

The uranium province comprehends a very large area, approximately 1,200 km^2, and the metamorphic foundation present in the region called gneissic-complex of Lagoa Real shows structural features where are found the albitites – uranium host rocks [14]. Therefore, it was necessary to limit the researched area prioritizing collection spots in the vicinities of cities, towns, villages and districts of the region. The field of study comprehends the towns of Caetité, Lagoa Real and a small part of the Livramento de Nossa Senhora Town (Fig. 1). The population from the three towns totalizes 102,444 inhabitants [15]. The climate is tropical, and it becomes subdivided into semi-arid and semi-humid according to the town, with temperatures that vary from 14.5 to 33°C [16]. Annual precipitation is approximately 800 mm with two very definite seasons: the dry season – from May to September; and a humid season from October to April , when it occurs 80% of the rains [17].

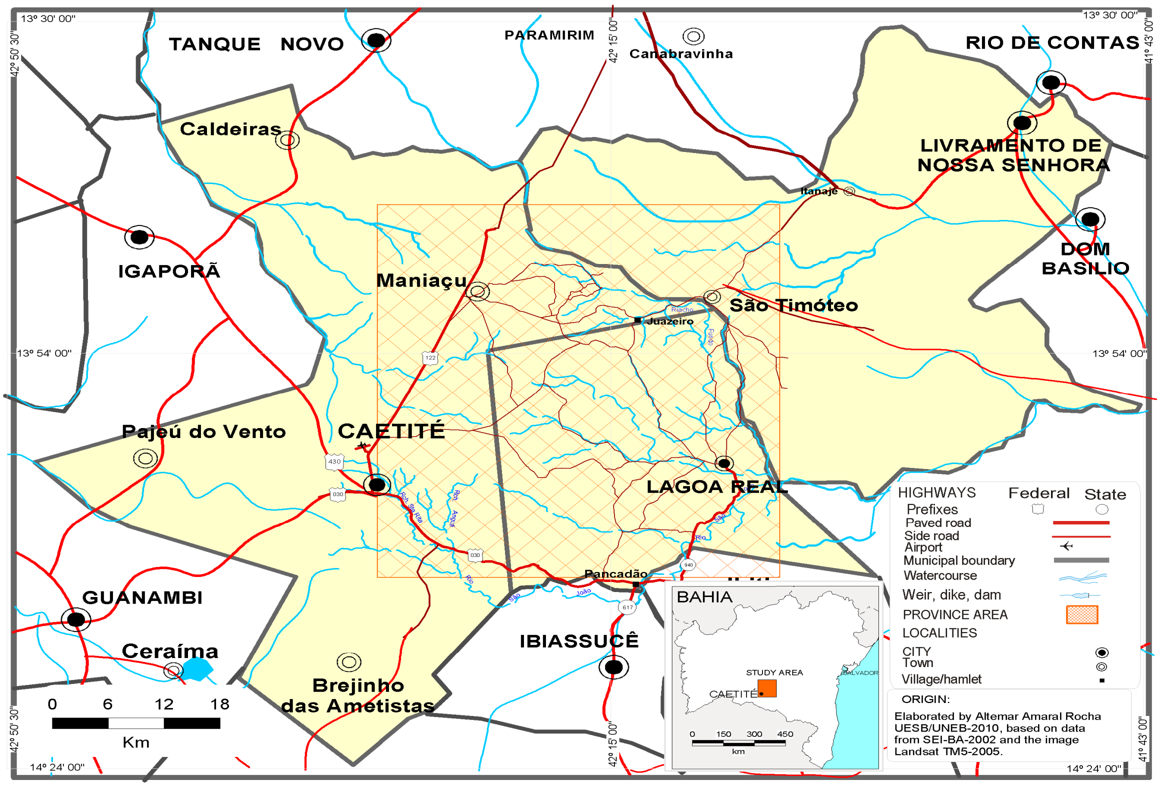

Figure 1. Caetité, Lagoa Real and Livramento de Nossa Senhora regions with some districts and villages, featured in the study area[18].

Geomorphologically, the region is situated in the Caetite massif (plateau), delimitated to north and south by the smoother surfaces of Paramirim and Cacule, respectively, totalizing an area of 1,200 km^2 containing 34 uranium deposits. In the restrict scope of uranium anomalies of the Lagoa Real Uranium Province emerges metassomatic metamorphic rocks of crystalline foundation with cataclastic structure notably granitoids, microcline-plagioclasic-augengneisses and albitites. These rocks are partially covered with dentritic sandy-clay sediments of tertiary-quaternary origin, a little or not consolidated at all, with thickness not over 50 meters [19, 20].

The urban population that lives in the proximities of the uranium province in the towns of Caetite, Lagoa Real and Livramento uses water from public supply which is treated and distributed. In the rural area water treatment for people's usage is almost inexistent, and due to long dry periods and the intermittence of the rivers in the region, the population obtains water from wells, cisterns, using small weirs, reservoirs and dams which are supplied during the rain [17]. The water originated from those sites is consumed most of the time without any treatment or radiometric verification even though it is a high background radiation region.

2. MATERIALS AND METHODS

During two consecutive periods, January and July 2010, in the towns of Caetité, Lagoa Real and Livramento de Nossa Senhora, a total of 32 points of superficial, underground and public supply water catchment were evaluated (Fig.2). In some homes it was also collected water from clay filters, originated from regional wells or the rain, which are filtered by the people themselves and stored in those recipients. The habit of filtering water using clay filters is very common in that region. Table 1 shows the sampling points characterized by their location and the source of water supply.

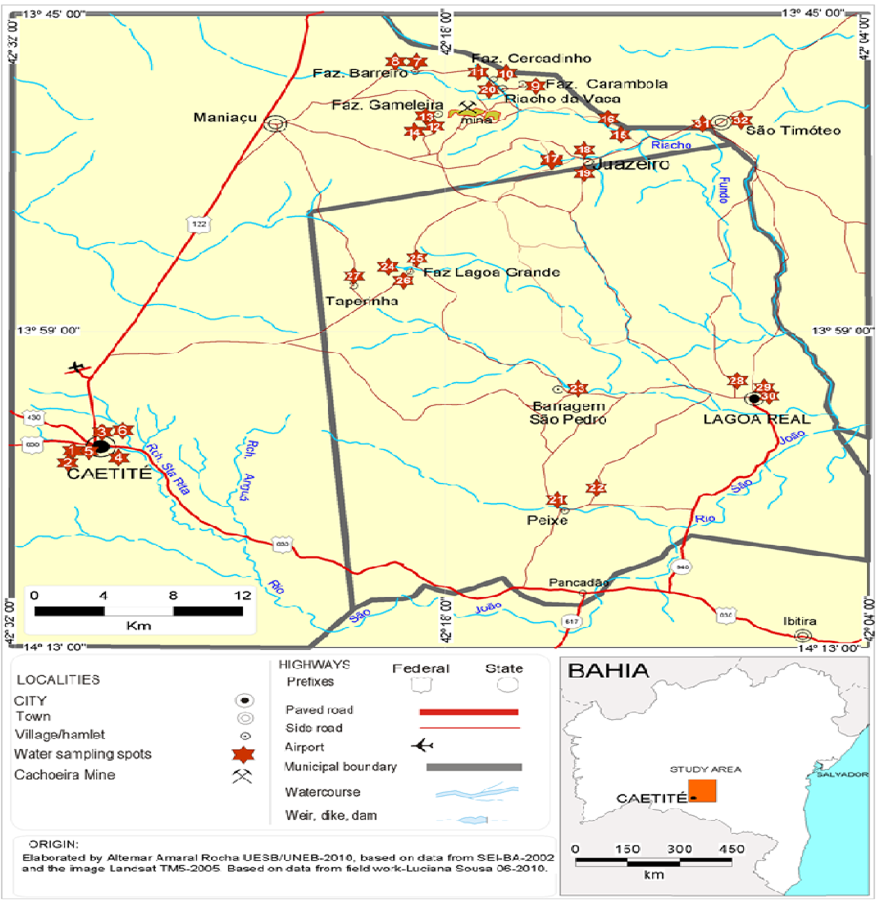

Figure 2. Points of water sampling in Caetité, Lagoa Real and Livramento de Nossa Senhora regions [21]

Table 1. Sampling points of water from several locations in central south Bahia state, Brazil

Municipality	Sampling Point	Location	Font of water supply
Caetité, BA	1	Urban Zone	public supply
	2	Urban Zone	public supply
	3	Urban Zone	public supply
	4	Urban Zone	public supply
	5	Urban Zone	public supply
	6	Urban Zone	well
	7	Rural Zone (Fazenda Barreiro - Maniaçu)	well
	8	Rural Zone (Fazenda Barreiro - Maniaçu)	water tank
	9	Rural Zone (Fazenda Carambola - Maniaçu)	well
	10	Rural Zone (Fazenda Cercadinho-Maniaçu)	well
	11	Rural Zone (Fazenda Cercadinho-Maniaçu)	rainwater
	12	Rural Zone (Fazenda Gameleira-Maniaçu)	Dam
	13	Rural Zone (Fazenda Gameleira-Maniaçu)	Dam
	14	Rural Zone (Fazenda Gameleira-Maniaçu)	rainwater
	15	Rural Zone	well
	16	Rural Zone	well
	17	Rural Zone (Juazeiro)	well
	18	Rural Zone (Juazeiro)(	well
	19	Rural Zone (Juazeiro)	well (filtered water)
	20	Rural Zone (Riacho da vaca)	well
Lagoa Real, BA	21	Rural Zone (Localidade Peixe)	well
	22	Rural Zone	well
	23	Rural Zone (Sao Pedro)	Dam
	24	Rural Zone (Lagoa Grande)	pond
	25	Rural Zone (Lagoa Grande)	well
	26	Rural Zone (Lagoa Grande)	well (filtered water)
	27	Rural Zone (Vila Taperinha de Lagoa Grande)	well
	28	Urban Zone	water before filtration
	29	Urban Zone	well
	30	Urban Zone	public supply
Livramento de Nossa Senhora, BA	31	Rural Zone (São Timóteo)	well
	32	Rural Zone (São Timóteo)	Housing

The water samples for the determination of the uranium concentrations were collected and stored in plastic recipients with a 45 mL capacity. After that the samples were acidified maintaining pH < 2 and submitted to refrigeration until the moment of the analysis. In the laboratory, the water samples were prepared by transferring 9 mL of the sample to a plastic tube of 15 mL together with 1 mL of indium, previously prepared, with 1 μg.L^{-1} concentration. To analyze the uranium concentrations it was used an inductively coupled plasma mass spectrometer (ICP-MS) Finnigan MAT, ELEMENT, (Bremen, Germany). Firstly, all samples were analyzed without filtration later on a few samples were filtered and re-analyzed. The ICP-MS calibration was done with uranium standard solutions, obtaining calibration four curves. The nitric acid used in the preparation of all standards is suprapure (distilled) in order to avoid possible interferences in the analyte reading due to high sensibility of the machine.

3. RESULTS

The samples were collected in two runs, the first one, in January 2010, corresponding to the rain season, and the second one in July 2010, corresponding to the dry season.

In the rain season, the uranium concentrations varied from 0.071 ± 0.005 μg.L^{-1} to 90 ± 1,5 μg.L^{-1}.

In the dry season, the uranium concentrations varied from 0.064 ± 0.005 μg.L^{-1} to 26.3 ± 0.3 μg.L^{-1}.

The results of the uranium concentrations for all sampling points, rain and dry season, unfiltered and filtered water, are summarized in Fig. 3.

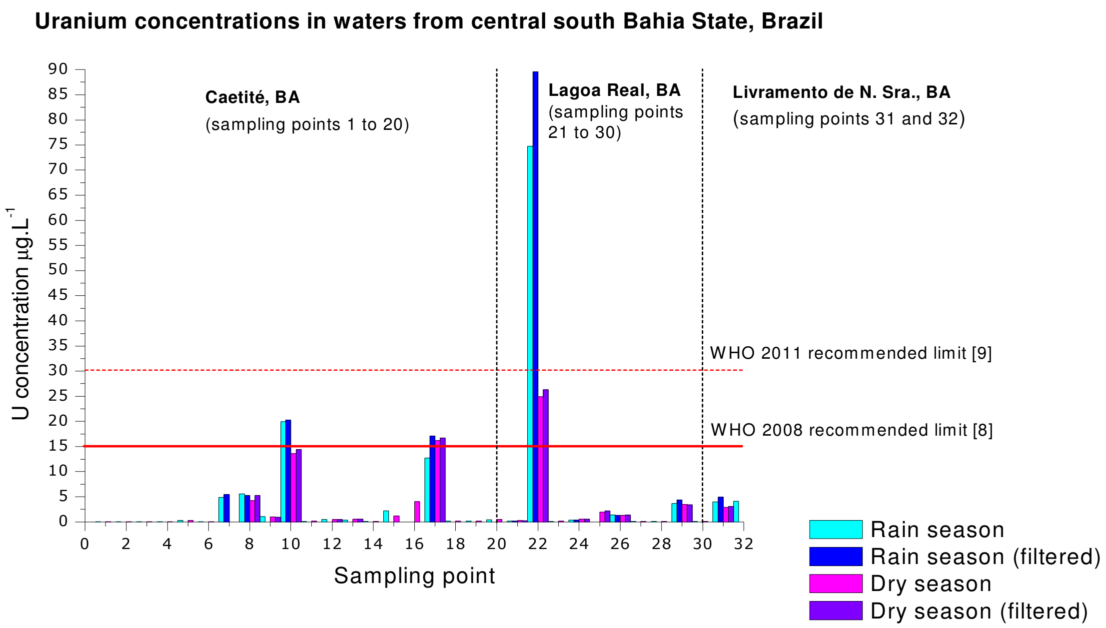

Figure 3. Uranium concentrations in μg.L^{-1} in surface, underground and drinking water of Caetité, Lagoa Real and Livramento de Nossa Senhora (Dashed horizontal lines represent WHO recommended guidance levels) [8,9]

4. CONCLUSIONS

The results show that the seasonality and the filtration process did not interfere significantly in the uranium concentrations results from the studied sites, except for sampling point 22, in the Lagoa Real region.

Almost all the results are below the guidance level of 15 $\mu g.L^{-1}U$ recommended by the World Health Organization in 2008 [8] and CONAMA [10].

In the Caetite region, sampling points 10 and 17, both of underground water in the rural zone, showed, respectively, uranium concentrations of 20.3 ± 0.3 $\mu g.L^{-1}$ and 17.1 ± 0.3 $\mu g.L^{-1}$. In the Lagoa Real region, for sampling point 22, underground water in the rural zone, the uranium concentrations in the dry season are 25 $\mu g.L^{-1}U$ and 26,3 ± 0.3 $\mu g.L^{-1}U$, respectively for the unfiltered and filtered sample. These results are higher than the guidance level of 15 $\mu g.L^{-1}U$ recommended by the World Health Organization in 2008 [8] and CONAMA [10]. Still, the results are so far lower than 30 $\mu g.L^{-1}U$, the current 2011 World Health Organization recommended guidance level [9].

Also for point 22, it was observed that, the uranium concentrations for the rain season are much greater than the ones for the dry season and there is also a difference between the unfiltered and filtered samples, 75 $\mu g.L^{-1}U$ and 90 $\mu g.L^{-1}U$, respectively.
Considering the WHO current recommended guidance level [9], only a single result, sampling point 22, in the region of Lagoa Real showed uranium concentrations pretty elevated for both rain and dry seasons, the maximum value registered being 90 $\mu g.L^{-1}U$ in the rain period. The same sampling of underground water also presented high concentrations of total alpha and beta radioactivity in a previous analysis [11].

The final evaluation of the results will be completed comparing the uranium concentration results with the total alpha and beta results.

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