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ASSESSMENT OF THE TOTAL URANIUM CONCENTRATION IN SURFACE AND UNDERGROUND WATER SAMPLES FROM THE CAETITÉ REGION, BA, BRAZIL

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ABSTRACT

At the region of Caetité, BA, it is located the largest uranium mine in exploration at present days in Brazil. During the uranium extraction process, it may be having an environmental contamination by this heavy metal due to rain water and other natural transport mechanism, with potential exposition risk to the local population. The aim of this work was to investigate the total uranium concentration in surface and underground water samples collected at the Caetité region, using the nuclear track registration technique (SSNTD) in a polycarbonate plastic. A 100 mL volume of water samples were initially treated in 10 mL of HNO_3 (PA) and concentrated by evaporation at a temperature around 80^{o}C. The resulting residue was diluted to a total volume of 25 mL without pass it to a filter. About 10 μL of this solution was deposited on the plastic detector surface (around 1.0 cm^2 area) together with 5 μL of a Cyastat® detergent solution (5%) and evaporated under an infrared lamp. All the resulting deposits of non volatile constituents were irradiated, together with a uranium standard sample, at the IPEN-IEA-R1 (3.5 MW) nuclear reactor for approximately 3 min. After irradiations, chemical etching of the plastic detectors was carried out at 60^{o}C, for 65 min. in a NaOH (6N) solution. The fission tracks were counted scanning all the deposit area of the polycarbonate plastic detector with a system consisting of an optical microscope together with a video camera and TV monitor. The average values of uranium concentrations obtained in this work ranged from (0.95 ± 0.19) $\mu g.L^{-1}$ to (25.60 ± 3.3) $\mu g.L^{-1}$. These results were compared to values reported in the literature for water samples from other regions and discussed in terms of safe limits recommended by WHO -World Health Oganization and CONAMA - Conselho Nacional do Meio Ambiente.

1. INTRODUCTION

In the last decades, a special attention has been given to the control of naturally occurring radioactive materials (NORM), in particular uranium, due to the increasing of activities mainly related to uranium ore milling, fertilizer production and coal thermal electric industries in several countries. The increasing of uranium presence in the environment is a matter of serious concern, because it can represent an important exposition source to the living organisms. In this way, the knowledge of uranium content in environmental and biological samples is important from the viewpoint of the radiological protection of the general population.

At Caetité County, south of Bahia state in Brazil, are located important ore mines mainly uranium, manganese, amethyst and iron. The largest uranium mine in exploration at present days is located in that County near the Maniaçu district. The region presents naturally high uranium concentrations in rocks and this geological characteristic together with the uranium milling activities may be producing a contamination of the water sources in that region.

The aim of this investigation is to report results from total uranium concentration determinations for surface and underground water samples collected in the Caetité County near a uranium mine, using the fission track registration technique (SSNTD).

2. EXPERIMENTAL METHOD

In order to determine the uranium concentration in the water samples it was employed the fission track registration technique in a polycarbonate plastic named PCLIGHT (l mm thickness) produced by Policarbonatos do Brasil S/A. The technique is simple, efficient and inexpensive, justifying its large use in several works reported in the literature worldwide in the last years. By employing a visual track counting system the efficiency of the technique is higher than 95%.

As control sample it was analyzed water samples collected from 4 rivers: Pitangueiras, Capim, Cachoeirinha e Olhos D'Água. These rivers are located in the Barretos County, north of the São Paulo state, in a region where uranium occurrence can be considered as being low ($< 1 \mu g.L^{-1}$). (GERALDO, et. al. 1979).

In the region of interest it was collected a tap water sample from the Maniaçu district, 3 underground water samples and 7 surface water samples, from the region near the uranium mine, as indicated in figure 1.

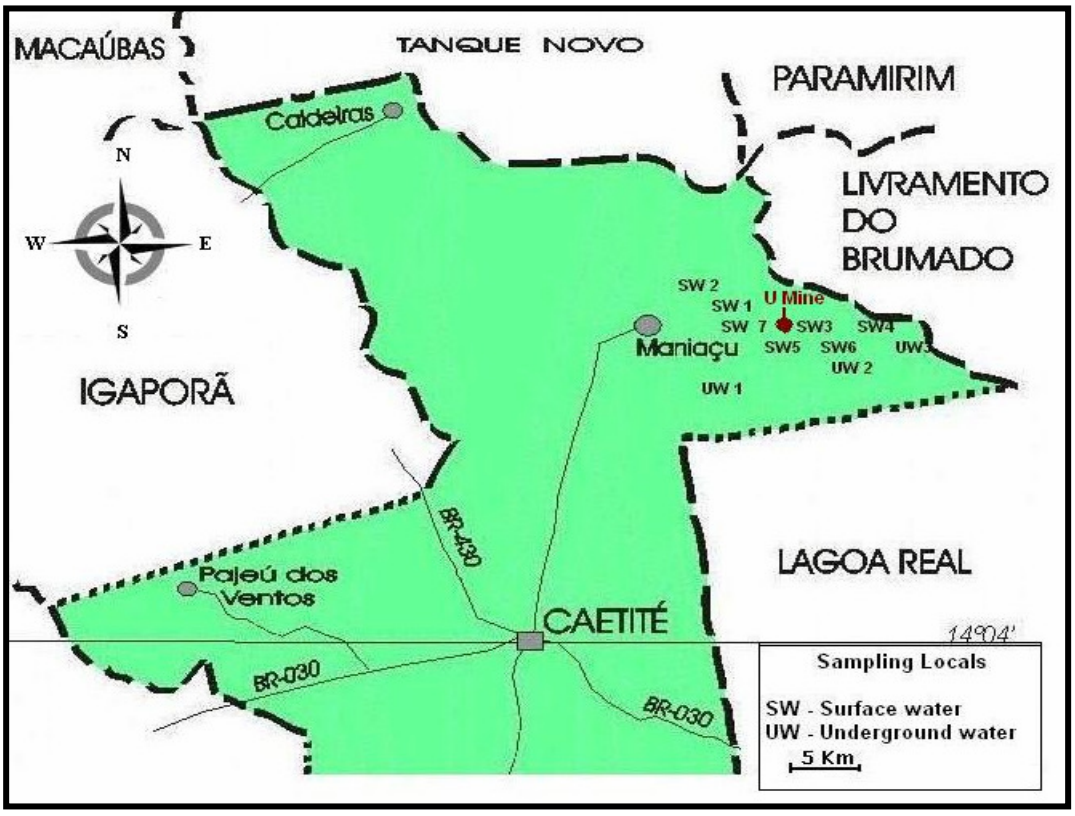

Figure 1: Map of Caetité (BA) region where are indicated the water sampling locals.

A volume of 100 mL was taken from each water sample and 10 mL of concentrated HNO_3 was added in order to acidify the sample to a pH lower than 2. In a thermal plate the solution was evaporated at a temperature of 80 ºC until getting a final volume of 25 mL. This procedure was carried out in order to obtain a homogeneous uranium distribution in the sample and a higher sensitivity for the technique. Aliquots of 10 µL of each solution were deposited on the plastic detector surface (approximately 1 cm^2 area) together with 5 µL of a Cyastat® detergent solution (5%, Cytec Indústrias) and evaporated under an infrared lamp (150 W) at a temperature around 70 ^oC. The Cyastat detergent solution has the function of an electrostatic neutralizer in such way that reduces the droplet surface tension and allows obtaining deposits with better homogeneity. In a similar way it was prepared deposits of a standard solution, having a known uranium concentration, to be employed as neutron flux monitor during the neutron irradiations.

All the plastic detectors containing the water and uranium standard sample deposits were wrapped with aluminum foil and piled inside an aluminum rabbit (22 mm diameter by 70 mm height) usually employed for neutron irradiations at IPEN-IEA-R1 (3.5 MW) pool type research nuclear reactor. The aluminum rabbit after being sealed by welding was placed for neutron irradiation near the reactor core in a local where the thermal neutron flux was around $1.2x10^{13}n/cm^2.s$. In this work, the irradiation time employed for all the samples was about 3 min. (GERALDO, et al. 2010)

After the irradiations, chemical etching of the plastic detectors was performed at a temperature of 61 ^oC, in a NaOH (6N) solution, for a time period of 65 min, to enlarge the fission tracks in order to be possible to observe them in a common optical microscope. The total track counting was obtained by scanning all the deposit area in the detector film with a binocular optical microscope coupled to a video camera and a TV monitor. As illustration, in figure 2 is presented fission tracks on an area of the plastic detector after chemical etching in the condition above cited. For each water and uranium sample it was carried out track counting at least for 5 deposit areas and the final value was determined by the average of the experimental results.

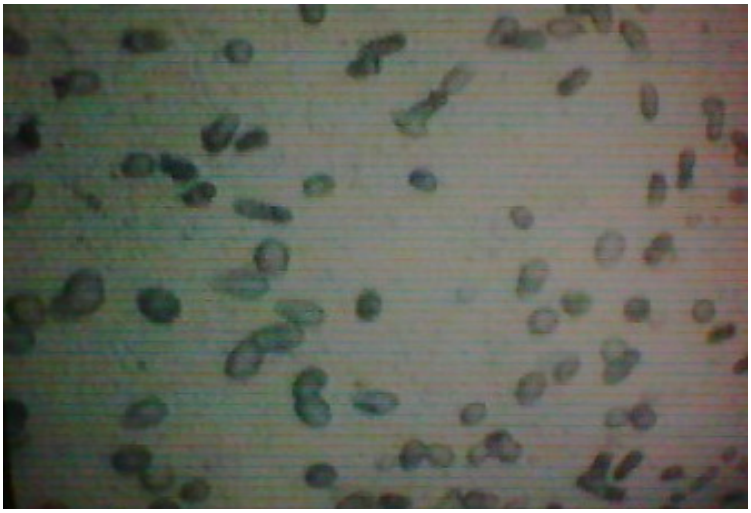

Figure 2: Fission tracks on an area of the PCLIGHT detector after chemical etching in the conditions adopted in this work.

3. RESULTS AND DISCUSSION.

The average values found for U concentration in control water samples from rivers where the uranium content is supposed to be low are listed in Table 1. The overall uncertainties for all the results obtained in this work were determined taking into consideration the following partial error sources: standard deviations of the mean values obtained in the track counting for the water sample and uranium standard sample deposits as well as the systematic error of 6.8% in the uranium content certified value for the standard uranium solution.
The calculated average value for the control water samples may be considered as representative of naturally occurring radioactive background for surface water systems (GERALDO et al., 1979; SCHMIDT, 2004; CARVALHO et al., 2007).
The average values obtained for surface (SW) and underground (UW) waters as well as for tap water from the Caetité County region are listed in Table 2. As it was expected underground waters presented the highest values. However, the values found for the surface waters are also much higher than those obtained for the control water even for the tap water which usually is very low. An interpretation for these abnormal occurrences of uranium background in the consumption waters are the presence of uranium ore mines in that region.
According to Brazilian agency for water quality control (CONAMA, 2005) the safe limit for uranium content in drinking water is 20 $\mu g.L^{-1}$. Recently the World Health Organization has recommended as safe limit in waters for drinking purposes 30 $\mu gU.L^{-1}$ (WHO, 2011). These limits are provisional guideline values and represent the uranium concentration that does not result in any significant risk to the public health. Taking into consideration the WHO recommendation all the water samples from the Caetité region seem to have uranium concentration values lower than the maximum limit. However, when compared with CONAMA limit the water from Quessengue Comunity Well is not appropriated for consumption**.**

Table 1: Average values for the uranium concentration in water samples from rivers of Barretos County region

Samples	U Concentration ($\mu g.L^{-1}$)
Cachoeirinha River	0.329 ± 0.089
Olhos d'água River	0.237 ± 0.054
Pitangueiras River	0.140 ± 0.038
Capim River	0.50 ± 0.11
Average Value	0.30 ± 0.15

Table 2: Average uranium concentrations in water samples collected in the Caetité County region.

Samples	U Concentration ($\mu g.L^{-1}$)
UW 1 - Tanquinho de Maniaçú Well	8.0 ± 1.5
UW 2 – Quessengue Comunity Well	25.60 ± 3.3
UW 3 - Lourinho Well	10.8 ± 1.5
SW 1 - Barreiro Lagoon	1.03 ± 0.31
SW 2 - Buracão Lagoon	1.78 ± 0.30
SW 3 - Joaquim de Ramiro Dam	2.26 ± 0.41
SW 4 - Colônia Lagoon	1.24 ± 0.23
SW 5 - Ind.Nac.Brasileira-INB Lagoon	1.30 ± 0.22
SW 6 - Quessengue Comunity Creek	2.22 ± 0.40
SW 7 – Ind.Nac.Brasileira-INB Creek	1.64 ± 0.33
Maniaçú Tap Water	0.95 ± 0.19

The uranium concentration interval obtained in this work for water samples from Caetité region are compared with those reported by other authors in Table 3. Some results were reported in $mBq.L^{-1}$ and to obtain the value in $\mu g.L^{-1}$ it was used the following conversion factor $1\mu g.L^{-1} = 12.35\ mBq.L^{-1}$ (GERALDO et al., 2010). As shown in this table, the interval of the present investigation agrees reasonably well with the ranges reported for Spain and India water samples. However, if it is considered only the surface waters analysis the present results are in better agreement with the other authors within the experimental uncertainties.

Table 3: Comparison of uranium concentration intervals reported in the literature worldwide for water samples.

Country	Water Type	Method	U Content ($\mu g.L^{-1}$)	Reference
Argentina	Bottled Mineral	Fluorimetric	0.04 – 11.0	Bomben et al. (1996)
Malaysia	Surface	Neutron Activation	0.33 – 1.40	Ramli et al. (2005)
France	Rivers	α Spectrometry	0.26 – 0,88	Schmidt (2004)
Slovenia	Surface	γ Ray Spectrometer	0.12 – 1,62	Korun and Kovačič (2011)
Portugal	Rivers	α Spectrometry	0.13 – 1.55	Carvalho et al. (2007)
Spain	Surface	α Spectrometry	1.62 – 21.1	Camacho et al. (2010)
India	Surface and Underground	Fission Track	0.9 – 63.0	Singh et al. (2009)
Brazil Caetité, BA	Surface and Underground	Fission Track	0.95 – 25.6	Present Work

4. CONCLUSIONS

Uranium concentration in surface and underground water samples collected at Caetité County, BA and control water samples from rivers of the Barretos County, SP have been measured in the present investigation. The average uranium concentration in control water samples is within the usual range found for consumption waters in several countries. However, for water samples from the Caetité region the uranium concentrations are much higher than the values found for the control waters. This may be interpreted as being a naturally occurring process since it is a region with high levels of uranium ore.

On comparing the present results with the provisional safe limit proposed by World Health Organization (WHO, 2011), all the waters analyzed in this work seem to have uranium concentration below the recommended level. However when compared with the maximum limit established by Brazilian agency for water quality control (CONAMA, 2005), one of the water samples (Quessengue Well) is not appropriated for public consumption.

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