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IMPACT OF ^{210}PB FROM OSAMU UTSUMI MINE ON SEDIMENT OF RIVERS IN CALDAS REGION, MINAS GERAIS

[1*]Pedro Henrique Dutra, [1]Maria Ângela de B. C. Menezes,**
[1*]Rubens Martins Moreira, [1***]Carlos Alberto de Carvalho Filho**

[1]Nuclear Technology Development Centre, Brazilian Commission for Nuclear Energy (CDTN/CNEN)
Caixa Postal 941, CEP 30161-970, Belo Horizonte, MG, Brazil;
* Post-Graduation in Radiation, Mineral and Material Science and Technology
phd@cdtn.br
** Division for Reactor and Analytical Techniques
menezes@cdtn.br
*** Division for Environment
rubens@cdtn.br
cacf@cdtn.br

ABSTRACT

The Osamu Utsumi Mine of the Indústrias Nucleares do Brasil (INB) is located in Caldas region, Minas Gerais, Brazil. It is a uranium mining and is in process of shutdown, decommissioning stage. CDTN/CNEN (Nuclear Technology Development Center, sponsored by Brazilian Commission for Nuclear Energy) is participating in this decommission step. One contribution will be the characterization of the environmental liability, determining the impact on the environment caused by mining activities. Several radionuclides are being analysed in diversified matrixes, however, this paper is about determination of ^{210}Pb in sediment of rivers. One reason to analyse ^{210}Pb is due to its long half-life (22.3 years) that may point out the carrier of ^{222}Rn, ^{226}Ra, ^{228}Ra, even U, in the region. Besides, it may be used to date sediment. The methodology applied to determine the ^{210}Pb activity in sediment was gamma spectrometry that was established at CDTN during the development of this study. The results confirmed the impact of the uranium mining on the environment.

1. INTRODUCTION

Lead (Pb) occurs naturally in the environment and has as isotopes ^{204}Pb, with natural abundance of 1.4%, ^{206}Pb, with 24.1%; ^{207}Pb with 22.1% and ^{208}Pb with 52.,4%. The last three ones also belong to the natural radioactive series, which begins with ^{238}U, ^{235}U and ^{232}Th, respectively, and occurs as final products of such decays.

Among the decay products of the natural radioactive series of ^{238}U, it is found ^{210}Pb, an important radioisotope from the point of view of the radiological protection, due to its half life of 22.3 years emitting β^- particles with energy of 61 keV and decaying to ^{210}Bi, emitting α of 3.7 MeV, decaying in the ^{206}Hg. Considering that it is a radioisotope of lead, it also presents the same characteristics and chemical toxicities.

Historically, ^{210}Pb began being detected indirectly by its son ^{210}Bi, or, by its descendant ^{210}Po. Studies developed later, suggested by Gäggeler et. al. 1976 [1], showed a new approach analyzing the ^{210}Pb itself by measuring the emitted gamma radiation of 46.5 keV. Such technique has been applied in order to detect ^{210}Pb, using scintillators and gamma spectrometers.

In areas where the minerals of uranium and thorium are explored, lead is present in their minerals due to its occurrence in the end of the chain of radioactive disintegration of those isotopes. In these areas, the main indicator of the presence of such elements is the radon exhalation.

The radioisotope ^{210}Pb has its origin mainly in uranium mines or mines that had uranium aggregated to the ore extracted. By having a half life of approximately 22.3 years, the studied radionuclide can be considered an important tracer in order to date the sediment and/or an indicator carrier of radon, radium and even uranium in the region. Studies emphasize the importance in the environmental area aiming at determining the concentration of ^{210}Pb [2].

A series of recent geochemical studies investigates the radioactive isotope ^{210}Pb, and showed its capacity of measuring the accumulation rate of sediments in the environment [3].

By using this concept of tracer and the indicator of radon carrier, radium and uranium due to the presence of ^{210}Pb in the environment, it is been developed a work on environmental analysis in an open uranium mine, Osamu Utsumi, together with the plant of extraction of ore and uranium which belongs to the Indústrias Nucleares do Brasil - INB (Brazilian Nuclear Industries) located in Caldas, a city in south of Minas Gerais State, which is facing a closing process [4].

At first, the mine environmental monitoring aimed at determining the long-lives radionuclides of the ^{238}U series, natural series of longer concentrations, once the presence of the sons of ^{232}Th would not be meaningful due to its low concentration in the area. Presently, both ^{238}U and ^{232}Th are monitored constantly, as well as other elements which have low contribution to the closing process that occurs in that area, to a better choice of the one that will be installed in order to conclude the (Plan for the Recovery of the Degradated Area) - Plano de Recuperação de Área Degradada (PRAD) [4].

This study aims at determining the activities of ^{210}Pb in samples of sediment of the rivers near the Osamu Utsumi Mine, focusing on the geoprocessing study of the region.

2. STUDY AREA

The studied area is located in Campo do Cercado, in the municipality of Caldas, in the central south region of the plateau of Poços de Caldas (PPC), in the southwest region of Minas Gerais State (Figure 1). It is located around 30 km from the cities of Caldas, Poços de Caldas, Águas da Prata and Andradas, and it is approximately 180 km from São Paulo, which is 350 km from Belo Horizonte and 360 km from Rio de Janeiro [5].

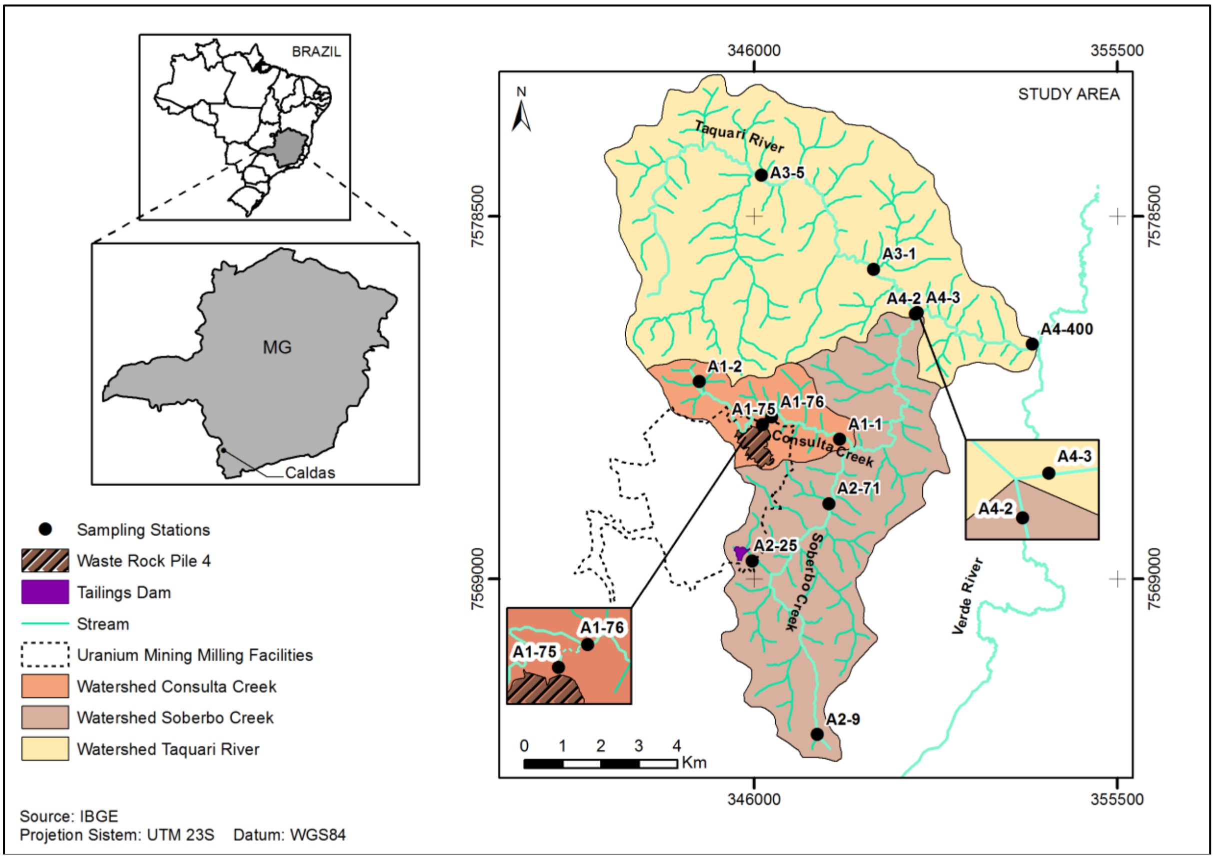

Figure 1: Map of the site of UTM-Caldas

From the point of view of hydrology, the Complex of INB is in Caldas on the watershed between two drainage systems: Verde and Antas [6].

Focusing on the project named INCT-Acqua [7], which evaluates the quality of water and sediments in the surrounding areas of the enterprise in Caldas, and that takes into account the focuses of potential contamination, it was defined the Study Area as shown in Figure 1. It is constituted by Taquari River Basin (affluent of the left riverside of Verde River), sub-divided by its three main sub-basins: stream of Consulta, stream of Soberbo and Taquari River. It can be observed the localization of the 12 stations where the samples of water and sediment were collected [5].

Considering the geology of the region, the Uraniferous District of Poços de Caldas was the only one in the world where uranium was minered in deposits related to alkaline rocks. The uranium field of Cercado occurs mainly in the form of uraninite, sometimes with nodular massive character, sometimes with a dust aspect. The oxide presents a high pureness level and contains impurities, such as, lead (± 1%), thorium (2%), zinc and iron (traces) [6].

3. METHODOLOGY

In this work, it was applied the gamma spectrometry technique in order to determine ^{210}Pb. It is a non destructive analysis.

3.1. Collecting and samples preparation

The samples analyzed in this work came from the collecting campaigns carried out in the end of the dry and rainy seasons of 2010 in the study area, distributed in 12 pre-determined sampling points [8]. They were collected with a "rock-islander" sampler in the riverbeds, sieved with a sieve of 2 mm (10 mesh) and stored.

At the Laboratory of Soil Samples and Sediments, building 3, Environmental Service-SEMAM, of the Nuclear Technology Development Center/National Commission for Nuclear Energy, CDTN/CNEN, the collected samples were dried at room temperature. The agglutinated grains were separated by using a proper roll which was put above the sample and had repeated and light movements, carrying out the process then. After that, the samples of raw material were removed in order to be analyzed. The objective was to determine ^{210}Pb in total samples. In the end, 24 samples were prepared to be analyzed.

3.2. Preparation of the samples to be analyzed

For the analysis by gamma spectrometry, a recipient of polyethylene with 67 mm of diameter by 27.8 mm height was chosen.

The samples were weighted in the recipients with range of 70 to 140 grams, trying then to fill the recipient in order to keep all the samples with the same height. The mass varied due to the variation of the density of the samples.

For this study, it was acquired a certified reference sample, IAEA-447, composed of a mixture of soil and moss, containing natural and artificial radionuclides. This material was supplied by the International Atomic Energy Agency (IAEA) (http://nucleus.iaea.org/rpst/ReferenceProducts/ReferenceMaterials/Radionuclides/IAEA-447.htm). Such material was used as an internal standard, then, it followed the same procedure for the samples. On 15th November 2009, the activity was (420 ± 20) Bq kg^{-1} and in the period of the analysis, June 2013, it was (397.36±19) Bq kg^{-1}.

3.3. Gamma Spectrometry

The spectrometer used at the Laboratory for Radiometrical Measurements, Reactor Service and Analytical Techniques – SERTA, of CDTN/CNEN, is composed of a hyper-pure germanium detector, CANBERRA, model GX5019, with related efficiency of 50% and a resolution of 1.9 keV for the gamma energy of 1.33 MeV from ^{60}Co. This detector is equipped with a carbon window, with 6.51cm of external diameter and associated electronics. The program Genie 2000, CANBERRA was used for the acquisition and analysis of the gamma spectra. Each sample was submitted to the gamma spectrometry for an enough period of time in order to achieve a counting statistics with uncertainty inferior to 10% under the same counting geometry.

4. RESULTS AND DISCUSSIONS

The period of the analysis of the 24 samples occurred between January and April. Each sample was counted in a specific time in function of the activity of ^{210}Pb in the sample. The results are shown in Table 1 and were calculated from the obtained spectra, focusing on the area of the gamma peak which interested.

Table 1. Results of the activities of ^{210}Pb of the samples

Collecting site	Collecting points	Rainy season Activity (Bq kg^{-1})	Dry season Activity (Bq kg^{-1})
Source of Consulta Stream – A1-2	1	214 ± 30	219 ± 27
Nestor Figueiredo Basin – A1-75	2	1226 ± 254	1791 ± 226
Source of Soberbo Stream – A2-9	3	113± 15	106 ± 14
Waste Basin – A2-25	4	2094 ± 256	1966 ± 246
Source of Taquari River –A3-5	5	131 ± 17	123 ± 17
River mouth of Taquari river – A4-400	6	311 ± 41	725 ± 91
After encounter Bacia Nestor Figueiredo – A1-76	7	316 ± 41	452 ± 58
River Mouth of Consulta Stream – A1-1	8	590 ± 76	545 ± 67
After meeting of the Waste Basin with Soberbo Stream – A2-71	9	272 ± 35	285 ± 36
Taquari Hill (Taquari River) – A3-1	10	242 ± 32	270 ± 35
Source of Soberbo Stream – A4-3	11	310 ± 40	367 ± 46
Encounter of Soberbo With Taquari River – A4-2	12	331 ± 42	327 ± 40

By analyzing the samples between the periods of seasonality, it was possible to check that the activity of ^{210}Pb kept certain constancy, and just a small alteration in its activity in each determined sampling point was observed, except in some occasions. Such small alteration can be seen in Figure 3, where the activities of ^{210}Pb are disposed in the collected samples, considering “R”, the ones collected in the rain season and with “D”, the ones collected in dry season.

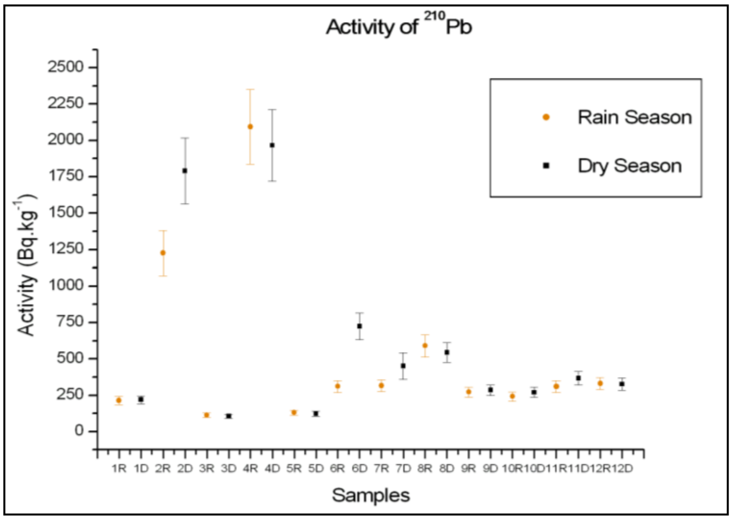

Figure 2. Comparison between the results in the samples collected in the rainy season and (R) dry season (D)

The results of sample 2, rainy and dry season, show an increase in the activity of ^{210}Pb in the dry season, suggesting then an influence of the sterile platforms, it is located above the sampling point, that is, they are disposed piles of aggregates of one or more minerals taken from a mine and have no value. [9]. This point is a place which receives water from these platforms. One explanation could be the accumulation of material retained during the rainy season.

The results of the activities at the point where sample 6 was collected, in the dry and rainy season, in which it is checked a higher variation between the seasons, suggests that an accumulation of ^{210}Pb in the dry season occurred.

Points 2 and 4, which have a higher activity of ^{210}Pb, are basins of control of the old uranium mine that are used to treat their waste and make the release of this water to the watercourse.

The sample of reference - IAEA-447 – was analyzed in quadruplicate, at the same geometry of the samples. Table 2 shows the activities obtained experimentally.

Table 2: Activity of ^{210}Pb in the Material of Reference used as an internal standard

Reference Material IAEA-447		
Sample	Experimental Values (Bq kg^{-1})	Certified Value, June 2013 (Bq kg^{-1})
D1	406 ± 52	397 ± 19
D2	422 ± 53	
D3	418 ± 52	
D4	418 ± 52	

Figure 3 shows Table 2 represented graphically to a better view of the activities obtained experimentally in comparison to the certified value.

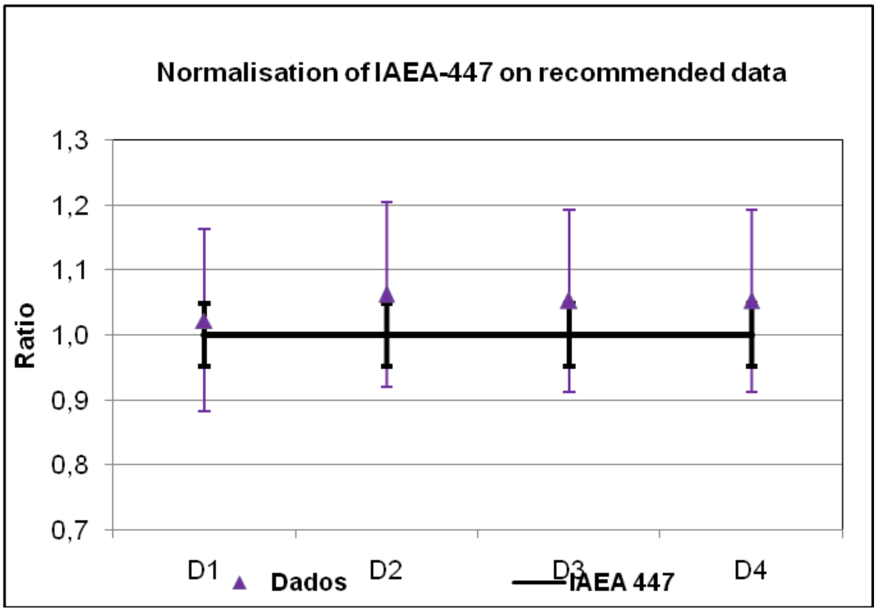

Figure 3. Ratio between the activities of Reference Material, IAEA 447, obtained experimentally by the certified value

Figures 4a, 4b show the values of the ^{210}Pb activities in the dry and rainy seasons, respectively, in all collecting points from the source and the river mouth of Consulta River.

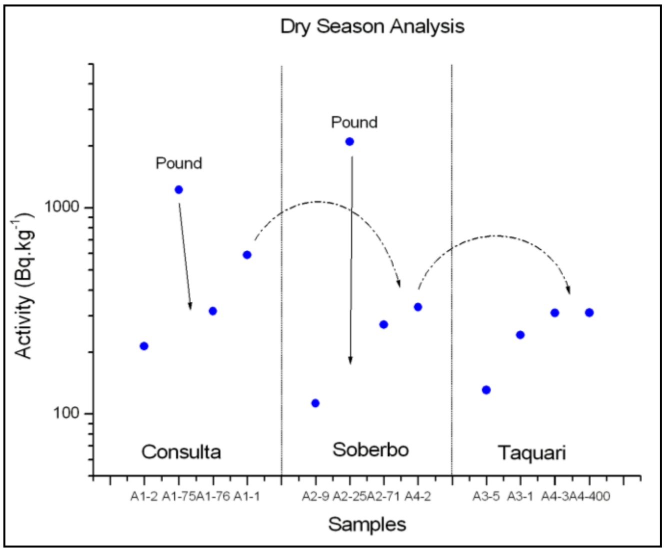

Figure 4a. Activity of ^{210}Pb in sediment samples of the collecting points in dry season

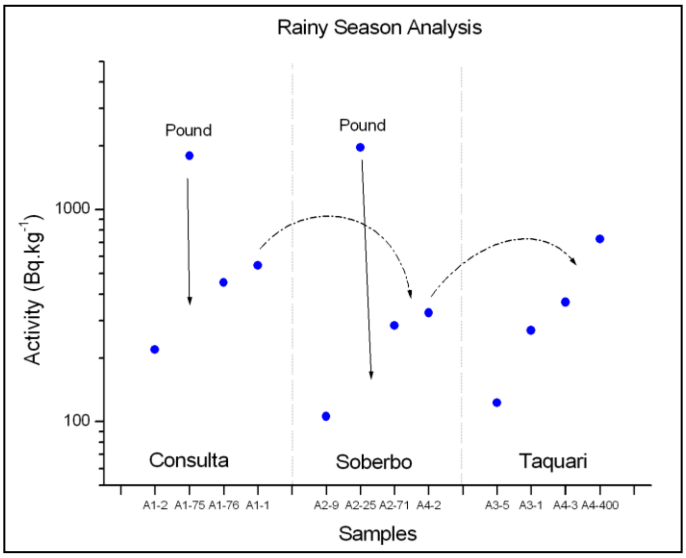

Figure 4b. Activity of ^{210}Pb in sediment samples in the collecting points in rainy season

The obtained results make possible to check the debouch of one river to the other is causing a gradual increase in the activity of ^{210}Pb in the river and a big increase of sediment in the bed of the river. Thus, the results suggest that the extraction of uranium from the mine caused an increase of ^{210}Pb in the region.

5. CONCLUSIONS

The results obtained from the activity of ^{210}Pb from the collected samples of sediment from the Osamu Utsumi Mine have influenced in the concentration of the ^{210}Pb activity in the rivers of the region. Moreover, in the dry season, there is an accumulation of sediment and because of that there is an accumulation of ^{210}Pb in the bed of the rivers. The results also show that from the river mouth of Consulta river, as in the dry season as in the rainy season, there is a significant increase in the ^{210}Pb activity, mainly after the debouche of a watercourse in another one.

The reference material IAEA-447, used as an internal standard showed that the applied gamma spectrometer procedure is in accordance with the measures and also that the obtained experimental data are in agreement with the certified values.

For future works, the analysis of the area near the watershed in order to check radiometric area of the region is suggested.

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