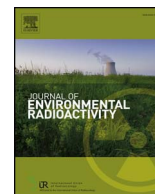




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Assessment of uranium release to the environment from a disabled uranium mine in Brazil

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ABSTRACT

The Ore Treatment Unit (in Portuguese Unidade de Tratamento de Minérios - UTM) located in Caldas, MG, Brazil is a disabled uranium mine. Environmental conditions generate acid drainage leaching metals and radionuclides from the waste rock pile. This drainage is treated to remove the heavy metals and radionuclides, before allowing the release of the effluent to the environment. To validate the treatment, samples of the released effluents were collected at the interface of the installation with the environment. Sampling was carried out from 2010 to 2015, and the activity concentration (AC, in Bq l^{-1}) of uranium in the liquid effluent was analyzed by arsenazo UV-Vis spectrophotometry of the soluble and particulate fractions, and of the sum of both fractions. Descriptive statistics, Z test and Pearson R^2 correlation among the fractions were performed. Then, the data were organized by year and both ANOVA and Tukey test were carried out to group the means by magnitude of AC. The annual mean ranged from 0.02 Bq l^{-1} in 2015 to 0.11 Bq l^{-1} in 2010. The soluble fraction showed a higher AC mean when compared to the mean of the particulate fraction and no correlation of the data could be observed. Concerning the magnitude of the release, the ANOVA associated with the Tukey test, identified three groups of annual means ($AC_{2010} > AC_{2011} = AC_{2012} = AC_{2013} = AC_{2014} > AC_{2015}$). The mean values of uranium release at the interface installation-environment checking point (point 014) were within the Authorized Annual Limit (AAL) set by the regulator (0.2 Bq l^{-1}) indicating compliance of treatment with the licensing established for the unit. Finally, the data showed a decreasing tendency of U release.

1. Introduction

The region of the Poços de Caldas plateau, state of Minas Gerais, Brazil, is considered with Naturally-Occurring Radioactive Materials (NORM). The first uranium mine in South America known as Unidade de Tratamento de Minério – UTM (Ore Treatment Unit), Caldas, is located there.

The discovery of uranium in the region dates back to 1948 and the mine was opened in 1971. Regular operation of the mine began in 1982 and was carried out for 13 years (Pedrobom, 2016). During the operation $1.08 \cdot 10^8$ t of sterile waste (include the whole exploited material) was moved to the waste rock pile (Nobrega, 2007) which discharged radioactive contaminated effluents over time.

In mines where rainfall is high the water balance is controlled by the weather and it is generally not possible to control the discharge of liquid effluents. In such cases, it is necessary to establish procedures and

criteria for releasing discharges, based on concentration limits for each contaminant and, when necessary, to use an effluent treatment technology to reduce the release values so that they remain within authorized limits (IAEA, 2002). This is the case of UTM.

At UTM, the sterile is able to generate Acid Mine Drainage (AMD); an average of $1.10 \cdot 10^5 \text{ m}^3 \text{ month}^{-1}$ of acid drains, with pH of 1–3.3, and uranium is generated at a concentration that can reach 10 mg l^{-1} ($\cong 240 \text{ Bq l}^{-1}$) of U_{nat} (Boniole, 2016).

Among these deposits of barren waste, waste rock pile 8, with an area of 64.4 ha and volume of $1.5 \cdot 10^7 \text{ m}^3$ calls attention to the Cercado stream (Fig. 2). Point 041, where the treated effluent enters into the Águas Claras Basin, is the main release point of this waste rock pile. This basin releases treated effluents to the environment, via point 014 and this point was chosen by the regulator as checking point for environmental impact monitoring (Pedrobom, 2016).

Some facilities are authorized to release effluents containing

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radionuclides and heavy metals during operation. Even after the end of the operation, this release may continue to occur. This is the case of UTM, which is authorized to release effluents containing U and other radionuclides (Pereira et al., 2013).

Currently, there are several international standards for the control of radionuclide AC in food and water. They are mainly related to public exposures and the requirements for such recommendations are comprised in the IAEA safety standards (IAEA, 2014). Specific criteria for responses to radiological and nuclear emergencies related to AC in water and food (IAEA, 2011 e 2015) and in emergency situations for drinking water (WHO, 2011) and for food in international trade (FAO and WHO, 2006) have been established in international regulations.

As part of the licensing of the facility, several studies on the release of effluents containing uranium and other radionuclides were carried out, evaluating the Radiological Environmental Impact Assessment (REIA) (Fernandes et al., 1994; Amaral, 1982, 1985, 1988, 1993) in terms of an increase in effective dose compromised due to practice, in agreement with the legislation at the time (CNEN, 1973). This approach was maintained in the subsequent legislation (CNEN, 1988) and in the current one (CNEN, 2014). Before UTM operation, the baseline of the region was established by Amaral's studies (Amaral, 1982, Amaral et al., 1985), and the mobility of radionuclides in the plateau was evaluated (Azevedo et al., 1988; Barcellos et al., 1990).

The facility is authorized by the regulator, CNEN – Brazilian National Commission for Nuclear Energy, to release effluents containing U_{nat} at the maximum concentration of 0.2 Bq l^{-1} at point 014 (Pereira et al., 2013) and the acid drains produce effluents with concentrations of up to 240 Bq l^{-1} (Boniole, 2016). A treatment system based on co-precipitation of the U with calcium hydroxide [$\text{Ca}(\text{OH})_2$] was designed to adapt the effluents to the operation permit for effluent release. This treatment generates 3000 T y^{-1} of calcium diuranate (Cipriani, 2002; Lima, 2014).

Aiming to evaluate the release of effluents by the installation, an environmental monitoring program was established, based on Amaral's study (1982).

The present study aims to evaluate the release, of treated effluents containing uranium from waste rock pile 8 to the environment (point 014) by UTM, over the years of 2010–2015, evaluating the adequacy of UTM to regulatory controls at the plant/environment interface.

2. Methodology

2.1. Study area

The study area is located in the city of Caldas, in the state of Minas Gerais, Brazil, as shown in Fig. 1. UTM possesses three effluent release

points, called point 014, 025 and 076 (see Fig. 2). In this study, we analyzed point 014, associated with the term source waste rock pile 8 that releases effluents to the Águas Claras Basin via point 041. The released material, after entering in the Águas Claras Basin, flows into the Cercado Stream, at point 014, one of the interfaces of the installation with the environment (Fig. 2).

2.2. Sampling and uranium analysis

Samples were collected when release occurred, generally once a week, in polypropylene flasks with a capacity of 1 L and screw cap. The vials, duly identified, were flushed with water from the collection site. Immediately after collection the samples were acidified with nitric acid in the proportion of 1 ml conc. HNO_3 per liter of water.

At the laboratory, the samples were filtered on cellulose acetate filters ($0.45 \mu\text{m}$). The filtrate, from now on called soluble fraction, was analyzed and the AC of uranium determined using the arzenazo UV-Vis spectrophotometry method following Savvin (1961; 1964). The material retained onto the filter, called the particulate fraction, underwent acid digestion ($\text{HNO}_3:\text{HPO}_4$, 30:1, v:v) at a temperature not exceeding 90°C and the uranium determined as above. The sum of the AC of the particulate and soluble fractions was considered to be the total release of U.

2.3. Statistical analysis

The data were submitted to descriptive statistics to evaluate means, standard deviations and number of analyzes performed (Rosner, 2000).

The mean uranium AC of the soluble and particulate fractions were compared by the Z test and the Pearson R^2 correlation test (Rosner, 2000). The Z test applied to soluble and particulate fractions used the following hypotheses:

$$\begin{aligned} H_0 &= \text{the evaluated means are identical} \\ H_1 &= \text{the evaluated means are different} \end{aligned}$$

The set of data organized by year was submitted to an ANOVA analysis using the values of total release to evaluate the existence of differences of AC along the years and to a Tukey test for ordering the annual means using the following hypotheses:

$$\begin{aligned} H_0 &= \text{the annual means studied are identical} \\ H_1 &= \text{there is at least one annual mean different from the other} \end{aligned}$$

Finally, the adequacy of observed mean total release to the value authorized by the regulator (annual mean for the total release of 0.2

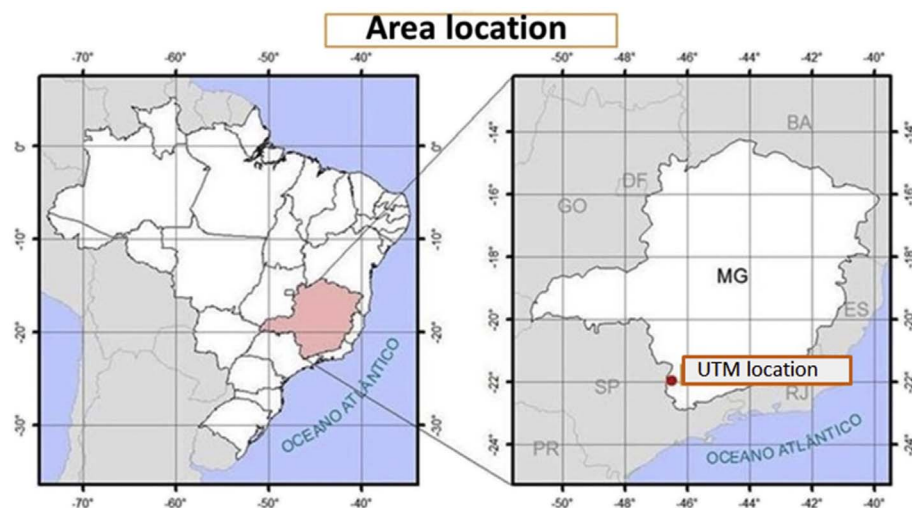


Fig. 1. UTM localization in Brazil.

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