

# **EVALUATION OF THE SEDIMENTATION RATE IN A SEDIMENT PROFILE OF BORTOLAN DAM – MG, BRAZIL**

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#### ABSTRACT

Volcanic extrusion is one of the main causes for the high levels of environmental radioactivity in the region of Poços de Caldas (MG, Brazil). Uranium mining during the 80's further increased the background radiation in the region, especially in the Ribeirão das Antas watershed, where Bortolan Dam is located. Besides the uranium mining facility operating in the region, other activities, such as agriculture, were also responsible for the enhancement of the levels of radioactivity. The dam sediment profile provides an appropriate compartment for the study of the temporal variation of these radionuclides and for the evaluation of anthropogenic contamination. The main aim of this study is to evaluate the sedimentation rate and the dating of a profile collected in Bortolan Dam, by using the geochronology method. The activity concentration of <sup>226</sup>Ra and <sup>210</sup>Pb were measured in the sediment profile by gamma spectrometry. The concentration of <sup>226</sup>Ra and <sup>210</sup>Pb varied from (217 ± 9) Bq kg<sup>-1</sup> to (286 ± 12) Bq kg<sup>-1</sup> and from (262 ± 11) Bq kg<sup>-1</sup> to (322 ± 34) Bq·kg<sup>-1</sup> respectively; and the sedimentation rates varied from 0.021 to 0.144 g cm<sup>-2</sup>y<sup>-1</sup>.

#### **1. INTRODUCTION**

The Poços de Caldas plateau was formed by the erosion of a volcano about 80 million years ago; it has an approximately circular shape, with a diameter of 35 km, an area of approximately 800 km<sup>2</sup> and altitudes between 1300 and 1600m. It is characterized by large circular volcanic depressions, called boilers, with high natural background radiation and a wide mineralogical and granulometric variation. In the 1980s, the extraction of uranium was started in the region of the Osamu Utsumi mine. This facility was operational until the year of 1995.

The uranium extraction activity and the extensive use of the soil for agricultural activities further increased the natural radiation levels in the region, including the water bodies and the sediments.

The region has two hydrographic basins: the Ribeirão das Antas basin and the Rio Verde basin, both tributaries of the Pardo River, inserted in the Rio Grande basin. The Ribeirão das Antas watershed occupies approximately 70% of the total area of the Poços de Caldas plateau. It has an area of about 455 km<sup>2</sup>; it is born on its southern border, at the altitude of

1640m and, in a course of 62 km, it transposes the northern border, in the Antas waterfall, at an altitude of 1180m [1]. The Ribeirão das Antas basin receives the effluents from the uranium extraction facility.

Several studies were concerned with the quality of water and sediments of the Ribeirão das Antas basin in Poços de Caldas [1, 2]. These studies emphasized that the artificial introduction of phosphate and nitrate (the primary elements in agricultural fertilizers), caused the pollution of rivers and springs and an ecological imbalance of algae and organisms, which decrease the oxygen content dissolved in water [1, 2, 3]. The Bortolan Dam, which is part of this basin, is considered a compartment that in the future may be used as a public water supply for the region; therefore, it is of interest to evaluate the temporal variation of the <sup>226</sup>Ra and <sup>210</sup>Pb in a sediment core collected in the dam and the evaluation of the sedimentation rate.

Records stored in natural archives, such as lake sediments, are used in a wide range of environmental programs, for example, in the assessment of changing erosion rates in a catchment arising from disturbances such as afforestation, deforestation, changing agricultural practice or to monitoring pollution by heavy metals, organic pollutants and other contaminants [4].

The climate in the area is marked by a rainy season from November to April and a dry season from May to October, when the rivers, ephemeral, tend to dry as they are fed by a smaller flow. In the rainy season, they present rapid floods fed by the high intensity of the rains. The aquatic system receives then loads of particles and elements that tend to decant in the bed of the water body, forming the natural deposits.

Among the <sup>238</sup>U decay series, the radionuclides <sup>226</sup>Ra and <sup>210</sup>Pb are considered as excellent geochronometers, since their isotopic signature allows dating the sediment profile age. This can be done by measuring the activity concentration of <sup>210</sup>Pb in the sediment profile and evaluating the difference between the <sup>210</sup>Pb activity from the atmospheric fallout, coming from the emanation of <sup>222</sup>Rn, and from the decay of <sup>238</sup>U itself in the soil [5].

Excess <sup>210</sup>Pb in the sediments over that in equilibrium with the *in situ* <sup>226</sup>Ra in sediments decays in accordance with the radioactive decay law,

$$C_{Pb} = C_{Pb}(\mathbf{0})\boldsymbol{e}^{-\lambda t} + C_{Ra} (1 - \boldsymbol{e}^{-\lambda t})$$
(1.1)

(where  $\lambda$  is the <sup>210</sup>Pb radioactive decay constant). This equation can be used to date the sediments, provided reliable estimates can be made of the initial <sup>210</sup>Pb activity  $C_{Pb}$  (0) in each sediment layer at the time of its formation [4].

This study aims to determine the concentration of <sup>226</sup>Ra and <sup>210</sup>Pb in core sediment collected in the Bortolan Dam and to evaluate the sedimentation rate and the dating of a profile through the geochronology, using the Constant Rate of Supply (CRS) model [6, 7]. The radionuclides activity concentration can be measured by gamma spectrometry, which has the advantage of being a non-destructive measurement that does not require leaching and radiochemical separation [8].

# 2. MATERIALS AND METHODS

The choice of the profile sampling location at the Bortolan Dam took into account the site of greatest sediment input and where there had been no disturbance, such as the presence of dredging. The geographical coordinates of the sampling location are  $21^{\circ}$  49' 07. 59" S and  $46^{\circ}$  38' 01.61" W.

Care was taken to select the zone of greatest depth of the channel to collect the maximum amount of sediment, and the procedure was performed on May 24, 2016, during the dry season of the region, to avoid the resuspension of sediment caused by precipitation.

The Figure 1 shows the dam environment. Dwellings and areas of cultivation, grazing and leisure occupy the region. Upstream, the site is occupied by industries such as the Companhia Brasileira de Alumínio (CBA).



Figure 1: Bortolan Dam.

The profile was collected by means of a gravity core sediment sampler. Figure 2 shows the moment when the sampler was launched from the boat, collecting the sediment with the least amount of interstitial water possible, to guarantee the integrity of the sample.





Figure 2: Gravity core sampler.

The sediment core was 18cm long and was sectioned every 2cm with the aid of a slicer and an extruder (7.2cm diameter), totalizing 9 samples. The slices were placed in aluminum containers, properly numbered and sealed, to avoid loss of humidity by evaporation (Figure 3).



Figure 3: Sectioning and pre-storage.

The sectioned samples were oven dried up to constant weight, at the temperature of  $60^{\circ}C \pm 5^{\circ}$ , and then the real humidity of each slice was obtained by equation 2.1, where  $m_u$  is the wet mass (initial mass of the sample – the mass of the container) and  $m_s$  is the dry mass (final mass – the mass of the container).

$$\mathscr{H}U = \left(\frac{m_u - m_s}{m_u}\right).100\tag{2.1}$$

Each sample was homogenized and a separate aliquot was used to determine the real density [9]. Using a pre-weighed pycnometer  $(m_p)$ , the sediment aliquot  $(m_s)$  was added, completing the remainder of the flask with distilled water, thereby obtaining the weight of  $m_t$ . The contents are discarded and a new weighing is carried out only with distilled water, providing the weight of  $m_{pa}$ .

The result of the real density is given by equation 2.2.

$$D = \frac{m_s}{(m_{pa} - m_p) - [m_t - (m_s + m_p)]}$$
(2.2)

These parameters are necessary for the determination of the total mass of solids of each fraction, used to obtain the corrected result of depth [10], sedimentation rate and sedimentation velocity. The following equation is used, where V is the volume of the sediment slice in cm<sup>3</sup>.

$$M_{s} = \frac{D.(1-U).V}{1+[U.(D-1)]}$$
(2.3)

The accumulated mass, calculated by the following equation, is obtained by dividing the total mass of solids by the surface area of the sample  $A_t$ .

$$M_A = \frac{M_S}{A_t} \tag{2.4}$$

The activity concentration of <sup>226</sup>Ra and <sup>210</sup>Pb in the sediment samples was performed by gamma spectrometry, by using a Canberra Hyperpure Germanium detector – HPGe, with 45% relative efficiency and lead shielding. The radionuclide <sup>226</sup>Ra was determined after 30 days, taking the average value of the photo peaks of their decay products: <sup>214</sup>Pb (295.21 keV and 351.92 keV). The <sup>210</sup>Pb was measured by the 46.50 keV photo peak. The self-absorption correction, due to attenuation of low energy gamma radiation by the sample itself, was not necessary, since the efficiency curve was obtained by measuring the reference material, with the same density as the sediment, in the same counting geometry. This curve was validated by measuring a certified standard reference material.

The efficiency curve was obtained by measuring the certified reference material IAEA - 447 [11] in the same sample geometry. For the spectral analysis, the Genie 2000 software was used.

# 3. RESULTS AND DISCUSSION

## 3.1. Real Humidity and Real Density

The humidity present in the samples derives from the interstitial water present between the grains and the sedimentary particles.

It should be noted that the decrease of humidity is one of the determining factors for the good integrity of the samples, because if there is an increase of it along the profile, this will be an indication that the area was disturbed or revolved through dredging or intense drought.

The real density was calculated by using the solids mass of each slice. The values presented linearity, demonstrating the increase of solids mass in relation to the amount of interstitial water, according to the humidity. These data are used to calculate and correct possible compactions of the profile by the water column.

Table 1 shows that the collected profile presents the decrease of humidity and increase of the solids mass in a regular way, validating its use for the study and showing that there are no major disturbances in the region.

Section	Real humidity (%)	eal humidity (%) Real density (g cm <sup>-3</sup> )	
1	67	2.4990	33.6306
2	68	2.5011	31.6303
3	58	2.4899	45.8265
4	61	2.7226	41.8746
5	57	2.6006	47.1662
7	56	2.5275	48.3558
8	55	2.5218	50.6112

#### Table 1: Real humidity, real density and mass of solids of profile

## 3.2. Radionuclides and Geochronology

In Table 2, the concentration values of the radionuclides for each section are presented, along with their uncertainties, calculated by error propagation. The values of atmospheric lead  $(^{210}Pb_{atm})$  are also shown.

Section	$^{226}$ Ra (Bq kg <sup>-1</sup> )	$^{210}$ Pb (Bq kg <sup>-1</sup> )	$^{210}$ Pb <sub>atm</sub> (Bq kg <sup>-1</sup> )
1	$230 \pm 10$	$322 \pm 34$	$92 \pm 35$
2	$217 \pm 9$	$288 \pm 24$	$71 \pm 26$
3	$222 \pm 9$	$286 \pm 29$	$64 \pm 30$
4	$255 \pm 10$	$317 \pm 25$	$62 \pm 27$
5	$277 \pm 11$	$280 \pm 29$	48 + 31
6	$252 \pm 9$	$262 \pm 10$	$10 \pm 13$
7	$286 \pm 12$	$298 \pm 32$	$12 \pm 34$

Table 2: Activity concentration of	<sup>226</sup> Ra,	<sup>210</sup> Pb and	<sup>210</sup> Pb <sub>atm</sub>	in the sediment	samples
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In reference [2], the <sup>226</sup>Ra and <sup>210</sup>Pb concentrations were determined in the surface sediments of two locations at the Bortolan dam. The results ranged from 180 to 280 Bq kg<sup>-1</sup>, for the concentration of <sup>226</sup>Ra, and from 220 to 500 Bq kg<sup>-1</sup>, for the concentration of <sup>210</sup>Pb. These results are of the same order of magnitude of the results obtained in this work, for the same radionuclides in the first section of the profile  $(230 \pm 10 \text{ Bq kg}^{-1} \text{ for } {}^{226}\text{Ra} \text{ and } 322 \pm 38 \text{ Bq kg}^{-1} \text{ for } {}^{210}\text{Pb}).$ 

Still in the reference [2], the values for <sup>226</sup>Ra and <sup>210</sup>Pb of surface sediments of other dams in the region were obtained, which are outside the area of influence of the Osamu Utsumi mine. These values are presented in Table 3.

# Table 3: Activity concentration of<sup>226</sup>Ra and<sup>210</sup>Pb in sediments samples from other dam<br/>in Planalto de Poços de Caldas [2]

	<sup>226</sup> Ra (Bq kg <sup>-1</sup> )			$^{210}$ Pb (Bq kg <sup>-1</sup> )		
	Point 1	Point 7	Point 11	Point 1	Point	Point 11
	(Cercado	(Cachoeirinha	(Cipó	(Cercado	7(Cachoeirinha	(Cipó
	Stream)	Stream)	Dam)	Stream)	Stream)	Dam)
Min	150	140	100	280	180	160
Max	390	220	320	500	260	640

The results found for the sediment samples from dams outside the area of influence of the mine, presented <sup>226</sup>Ra concentration ranging from 100 to 390 Bq kg<sup>-1</sup>, and for <sup>210</sup>Pb, ranging from 160 to 640 Bq kg<sup>-1</sup>. These concentrations are of the same order of magnitude

as those obtained in this work for the surface sediments  $(230 \pm 10 \text{ Bq kg}^{-1} \text{ for the } {}^{226}\text{Ra} \text{ and } 322 \pm 34 \text{ Bq kg}^{-1} \text{ for the } {}^{210}\text{Pb}).$ 

The results obtained for the concentration of  $^{226}$ Ra and  $^{210}$ Pb along the profile are presented in Figure 4, for both the real depth and the corrected depth. It is noticed that there is a decrease in the concentration of  $^{210}$ Pb along the profile, until reaching values of the same order of magnitude of the  $^{226}$ Ra, in the depth of 10 cm for the real depth and 4cm for the corrected depth. From these depths on, the two radionuclides are in equilibrium, indicating that there is no more deposition of the atmospheric lead ( $^{210}$ Pb<sub>atm</sub>).



Figure 4: Variation of Ra and Pb activity concentration with depth

The real depth was used for the determination of dating and the corrected depth was used for the determination of the sedimentation rate. The age of each section of the profile was evaluated by using the CRS model and the <sup>210</sup>Pb<sub>atm</sub> values. Using the  $\chi^2$  method, the function was adjusted to calculate the ages with the best fit line (figure 5).



Figure 5: Ratio between age and depth of profile

The sedimentation rate was obtained from the accumulated mass and age of each section (Table 4). A small sedimentation rate at the location becomes evident, ranging from 0.021 to 0.144 g cm<sup>-2</sup> y<sup>-1</sup> and a constant sedimentation velocity of 0.113 cm y<sup>-1</sup>.

Section	Sedimentation rate $g \text{ cm}^{-2} \text{ y}^{-1}$		
1	0.144		
2	0.070		
3	0.059		
4	0.039		
5	0.034		
7	0.024		
8	0.021		

 Table 4: Sedimentation rate of profile

It can be seen that the length of the sediment core, obtained by using the corrected depth, was 8 cm. This value does not agree with the real length of the core, that was measured when the core was collected, that is 18 cm. The determination of the age of the Represa Bortolan, obtained by using the real depth, gave as result 59 years. This result is in agreement with the effective age of the dam, since it was built 58 years ago.

#### 4. CONCLUSIONS

In the present work, it was observed that the concentration of radionuclides studied in the sediment profile of Bortolan Dam varied from  $217 \pm 9$  to  $286 \pm 12$  Bq·kg<sup>-1</sup> for <sup>226</sup>Ra and from  $262 \pm 10$  to  $322 \pm 34$  Bq·kg<sup>-1</sup> for <sup>210</sup>Pb. These values are of the same order of magnitude of the natural radiation of dam sediments, which are outside the influence of the uranium mining facility, showing that there is no measurable contribution of natural radionuclides in the sampling collection. The fact that all the locations mentioned in this work have a similar radionuclides concentration is justified by the geological conformation of the Poços de Caldas Plateau, which is classified as a radioactive anomaly, and the hydrology of Ribeirão das Antas, which tends to increase the concentration of radionuclides in the sediments.

It is necessary to emphasize that the Brazilian federal legislation does not establish limits for the concentration of radionuclides in sediments; therefore it is important to have a temporal record of the variation of the radionuclides concentration, in order to assess possible anthropogenic contamination.

A higher sedimentation rate and sedimentation velocity was expected, since these values are related to the contribution of debris coming from anthropogenic actions at the site. The values obtained were lower than expected, maybe due to the profile location, of difficult access and downstream of the dam, where the particles lose energy until their sedimentation and also due to the conformation of the dam, which presents meanders that can retain these particles. Based on this, a new site for collection will be studied.

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