

## **NORM ASSESSMENT IN WATER TREATMENT SYSTEMS/POÇOS DE CALDAS -BR CASE**

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### **ABSTRACT**

NORM is the acronym used to refer to naturally occurring radioactive materials. Besides being objects of study and monitoring such materials can be used as raw material or as by-products or waste of industrial activities. Oil and gas, mining and water treatment are examples of facilities that can handle NORM. In such cases, their concentration at significant levels from the perspective of environmental and occupational radiation protection may occur. This study aims to evaluate the presence of the natural radioactive <sup>238</sup>U and <sup>232</sup>Th series in the treatment of city water elements Poços de Caldas - MG (water, materials and waste). The study can serve as an indication of the necessity of a more detailed review in the locally and in the country on this radiological issue.

### **1. INTRODUCTION**

Naturally-occurring radioactive material (NORM) is the term used to describe materials containing radionuclides existent in the environment. Long-lived radioactive elements of include uranium, thorium and potassium, and any of their radioactive decay products, such as radium and radon (usually called their "daughters").

Mankind have always been in contact with ionizing radiation, as the natural radionuclides are present in the terrestrial biosphere, distributed along water bodies, biota, soil or even air (due to reactions that occur naturally) [1].

Once present in Earth's crust, these radionuclides can be found in some minerals and, therefore, be a component of the raw material, byproducts or residues of several industrial activities. Classic examples are mining and oil industries, as well as water treatment. Along the production process of those activities, the radioactive materials may undergo a change in concentration, being diluted or concentrated [2]. So, from the point of view of occupational and environmental radiation protection, monitoring and management of NORM within the context of certain industries becomes of great importance [3].

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Water is one of the most valuable natural resources. Especially in the face of climate change and population growth, abundant and clean water supply is a major challenge for modern civilization. Some level of radioactivity may be present in water supplies as a result of natural processes or from the disposal of radioactive materials [2]. Therefore, water treatment plants have been considered by several authors like NORM industries [4].

The Poços de Caldas Plateau presents what is called "radioactive anomalies", which are regions with natural radioactivity levels above those usually observed at the Earth's surface. A large hydrographic system permeates through the anomalous areas and also the region has high index of rain occurrence. These anomalies, not surprisingly, host a uranium mineralization which, in the past, was explored as the first Brazilian uranium mining. Now in decommissioning process, the facility has waste rock piles where there is the occurrence of acid mine drainage [5, 6].

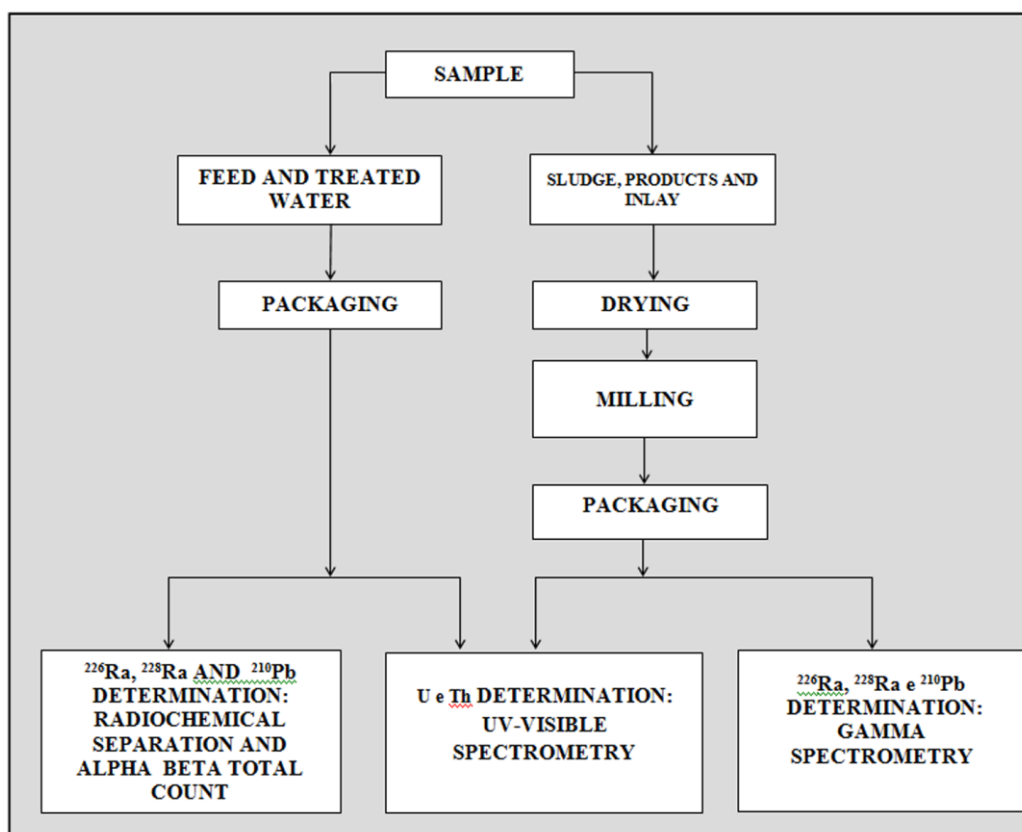
Taking into account the characteristics of the region and the main aspects mentioned above, according to Palomo et al. [7], the local water treatment plants in operation can lead to production of radioactive waste as, for instance, sludge with accumulated radionuclides or contaminated filters.

In Brazil, in addition to employing the most conventional treatment stations supply, for a long time the waste generated in water treatment plants are being discarded directly into water bodies without any kind of proper treatment [8]. Then, it becomes essential, especially in the region cited, a study aiming to assess the quality of the public water supply as well as of the waste generated in the treatment process.

## **2. EXPERIMENTAL**

### **2.1 Sampling**

Feed and treated water, input materials (aluminum sulphate and calcium hydroxide), scale and sludge were sampled in the local water treatment system for public supply. Feed water samples were collected in the feeding pipes prior to the beginning of the treatment, and the treated water sample were collected at the exit of the process. Sludge samples were collected directly from decanters when, by automatic processes, these were drained for cleaning. The inputs were collected before the mixture for the treatment and the scale was scraped from the walls of the decanters. The flow sheet below (Fig. 1) presents the treatment procedure of the samples prior to chemical and radiochemical analysis.



**Figure 1. Flowchart of the processing and analysis of samples.**

## 2.2 Analytical Methods

### 2.2.1 Gamma Spectrometry

Gamma spectrometry is an instrumental technique that can be used for the determination of a wide variety of radionuclides in different matrices. The potential of this technique allows the study of gamma emitters in energies that can range from 30 keV up to 2000 keV. In the present study, the technique of gamma spectrometry was used for the determination of radionuclides  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  and  $^{210}\text{Pb}$  using a Canberra Range detector model GX4510.

### 2.2.2 UV- Visible Spectrometry

The method used for determination of Thorium, which is extracted from aqueous solution using a solution of TOPO (Trioctylphosphine oxide) diluted in cyclohexane, followed by re-extraction in aqueous solution. On determination of uranium, the method is based on separation by extraction with tri-n-butyl phosphate (TBP) in a solution containing  $\text{Al}(\text{NO}_3)_3$ , EDTA and tartaric acid. The uranium stripping is then carried out with a solution of Arsenazo (III). Both were read from the Varian Spectrophotometer, model Cary 50.

### 2.2.3 Radiochemical Separation and Alpha Beta Total Count

This method is based on a chemical separation of radio and lead of the other elements present in a sample by selective precipitation. The Alpha and beta counting of the precipitates were carried out using a gaseous flow proportional counter Model S5-XLB Tennelec Canberra.

## 3. RESULTS AND DISCUSSION

The analytical results obtained for feed and treated water from the first sampling campaign are presented in Table 1, while the second campaign results shown in Table 2. For comparison of the results of the alpha and beta activity, it was considered the Ordinance n° 2,914 the Ministry of Health, December 12<sup>th</sup> 2011 [9], which provides directives for the control and monitoring procedures of the quality of water intended for human consumption.

**Table 1. Analytical results of feed and treated water samples (first sampling campaign).**

Water Treatment Plant	Source	<sup>226</sup> Ra (Bq.L <sup>-1</sup> )	<sup>228</sup> Ra (Bq.L <sup>-1</sup> )	<sup>210</sup> Pb (Bq.L <sup>-1</sup> )	U (Bq.L <sup>-1</sup> )	Th (Bq.L <sup>-1</sup> )	α-total (Bq.L <sup>-1</sup> )	β-total (Bq.L <sup>-1</sup> )
A	feed water	< 0,02	< 0,02	< 0,02	< 0,18	< 0,01	< 0,06	< 0,06
	treated water	< 0,02	< 0,02	< 0,02	< 0,18	< 0,01	< 0,06	< 0,06
B	feed water	< 0,02	< 0,02	< 0,02	< 0,18	< 0,01	< 0,06	< 0,06
	treated water	< 0,02	< 0,02	< 0,02	< 0,18	< 0,01	< 0,06	< 0,06
C	feed water	< 0,02	< 0,02	< 0,02	< 0,18	< 0,01	< 0,09	< 0,09
	treated water	< 0,02	< 0,02	< 0,02	< 0,18	< 0,01	< 0,06	< 0,06

**Table 2. Analytical results of feed and treated water samples (second sampling campaign).**

Water Treatment Plant	Source	$^{226}\text{Ra}$ (Bq.L <sup>-1</sup> )	$^{228}\text{Ra}$ (Bq.L <sup>-1</sup> )	$^{210}\text{Pb}$ (Bq.L <sup>-1</sup> )	U (Bq.L <sup>-1</sup> )	Th (Bq.L <sup>-1</sup> )	$\alpha$ -total (Bq.L <sup>-1</sup> )	$\beta$ -total (Bq.L <sup>-1</sup> )
A	feed water	< 0,02	< 0,02	< 0,02	< 0,02	< 0,01	< 0,48	< 0,65
	treated water	< 0,02	< 0,02	< 0,02	< 0,02	< 0,01	< 0,48	< 0,65
B	feed water	< 0,02	< 0,02	< 0,02	< 0,10	< 0,01	< 0,48	< 0,65
	treated water	< 0,02	< 0,02	< 0,02	< 0,05	< 0,01	< 0,48	< 0,65
C	feed water	< 0,02	< 0,02	< 0,02	< 0,20	< 0,01	< 0,48	< 0,65
	treated water	< 0,02	< 0,02	< 0,02	< 0,02	< 0,01	< 0,48	< 0,65

The results showed that, in the two campaigns, the total  $\alpha$  and  $\beta$  counts do not exceed the established limits of 0.5 Bq L<sup>-1</sup> and 1 Bq L<sup>-1</sup>, respectively. Article 38 of the Ordinance cited above, defines that, should the screening levels quoted are overcome, specific analysis must be performed for the radionuclides present and the results should be compared with stated references (in the same document). It is noticed that, given the reference levels of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ , 1 Bq L<sup>-1</sup> and 0.1 Bq L<sup>-1</sup>, the results obtained for samples of treated water, fit within established parameters as the activity concentrations detected are smaller than 0.02 Bq L<sup>-1</sup>.

The results  $^{210}\text{Pb}$  are also smaller than 0.02 Bq L<sup>-1</sup> in all treatment plants. It is worth mentioning that in spite of not being cited in the Ordinance, this radionuclide is also important from the point of view of radiological protection, because it could contribute significantly to the dose in a scenario of internal contamination [10]. The low values of concentration of U and Th are expected, once the total alpha and beta counts are within established parameters.

The composition of sludge produced during the treatment depends on the physical and chemical nature of the water and the sort and dosage of chemicals used [11]. There is no reference level available in the literature. However, the sludge produced (Table 4) in the treatment plants seems to pose no threat, in terms of environmental radioprotection, as they have the same order of magnitude of sediment samples of surrounding areas (Table 4).

**Table 3. Analytical results of sludge samples (first campaign).**

Sludge	$^{226}\text{Ra}$ (Bq.kg <sup>-1</sup> )	$^{228}\text{Ra}$ (Bq.kg <sup>-1</sup> )	$^{210}\text{Pb}$ (Bq.kg <sup>-1</sup> )	U (Bq.kg <sup>-1</sup> )	Th (Bq.kg <sup>-1</sup> )
A	198,00 ± 50	179,00 ± 9,0	145,00 ± 7,00	211,19 ± 7,92	214,92 ± 23,46
B	184,00 ± 46,00	156,00 ± 8,00	151,00 ± 8,00	475,30 ± 3,03	130,06 ± 14,56
C	183,00 ± 46,00	174,00 ± 9,00	209,00 ± 1,00	702,83 ± 8,88	210,00 ± 21,00

**Table 4. Mean values for sediments sampled in water treatment plants A and C surroundings [12].**

Sediment	<sup>226</sup> Ra (Bq.kg <sup>-1</sup> )	<sup>228</sup> Ra (Bq.kg <sup>-1</sup> )	<sup>210</sup> Pb (Bq.kg <sup>-1</sup> )	U (Bq.kg <sup>-1</sup> )	Th (Bq.kg <sup>-1</sup> )
A	110	226	144	161	228
C	214	293	350	379	298

The results obtained in the scales derived from the water treatment plant A show concentration of activities practically negligible, as well as in the B one. C's results show higher levels as shown in Table 4. Table 5 shows the results obtained for the scales from the second sampling campaign. Although the results from A and B treatment plants are very similar to the ones from the first sampling campaign, the C results showed a significant decrease. There is no conclusive explanation about this behavior yet. This may be explained by the results of the next sampling campaigns to be promoted.

Values found for U and Th may be considered low but, again, there is a difference between the results of the sampling campaigns. Again, some explanation about it is expected with the continuations of the studies.

**Table 5. Analytical results of the scale samples (first sampling campaign).**

Scale	<sup>226</sup> Ra (Bq.kg <sup>-1</sup> )	<sup>228</sup> Ra (Bq.kg <sup>-1</sup> )	<sup>210</sup> Pb (Bq.kg <sup>-1</sup> )	U (Bq.kg <sup>-1</sup> )	Th (Bq.kg <sup>-1</sup> )
A	69,00 ± 17,00	71,00 ± 4,00	37 ± 2,00	< 505,63	< 202,20
B	164,00 ± 41,00	226,00 ± 11,00	30,00 ± 2,00	< 505,63	< 202,20
C	1.192,00 ± 298,00	1.704,00 ± 85,00	301,00 ± 15,00	< 505,63	< 202,20

**Table 6. Analytical results of the scale samples (second sampling campaign).**

Scale	<sup>226</sup> Ra (Bq.kg <sup>-1</sup> )	<sup>228</sup> Ra (Bq.kg <sup>-1</sup> )	<sup>210</sup> Pb (Bq.kg <sup>-1</sup> )	U (Bq.kg <sup>-1</sup> )	Th (Bq.kg <sup>-1</sup> )
A	69,00 ± 23,00	105,4 ± 9,50	66 ± 20	< 244,00	134,00
B	164,00 ± 41,00	252,00 ± 18,00	74,00 ± 19,00	< 244,00	< 103
C	619,00 ± 110	790,00 ± 52,00	98,00 ± 24,00	< 244,00	170,00

### 3. CONCLUSION

The results presented here show that no significant presence of radionuclides was not detected in water provided to the population of Poços de Caldas municipality.

The main waste water treatment, sludge, also does not present radionuclide concentration high or significantly different from the sediments in the region of the the water treatment plants and surroundings. Possibly, Ra and Pb are being concentrated in the scales present in treatment tanks by means of manganese co-precipitation.

The results presented here are still to be considered preliminary, since the work is still being developed. It is known that the region has well defined climatic stations, as well as an intense rain period. We intend to continue to evaluate the treatment over a year to avail possible seasonal effects on the results.

Once completed, the presence of radionuclides in the scales of the treatment tanks will be evaluated regarding possible occupational and environmental radiation protection issues, in cooperation with the local water treatment operator.

### 4. REFERENCES

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