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Natural and artificial nuclides in Salesópolis reservoir

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ABSTRACT

Natural radioactivity is ubiquitous in the environment mainly due to the presence of the nuclides from the uranium and thorium series and ^{40}K. Although in the South Hemisphere nuclear tests have been fewer in number than that in the North, artificial radionuclides can also be found spread at ground level. In this study, the activity concentrations of natural nuclides from the uranium and thorium series, ^{40}K and the artificial ^{137}Cs were determined in a sediment core with 42 cm depth collected in the middle of the Salesópolis reservoir, located in the Metropolitan Region of São Paulo city (SPMR). The Usina Parque Rio Tietê reservoir belongs to the Alto do Tietê system for the capture, storage and treatment of water for SPMR. Therefore, the quality of the water and sediments of this dam is of great importance. The activity concentrations were measured by gamma spectrometry. Samples were measured and saved at regular intervals at a maximum of 160 000 seconds. The gross area were determined for each peak and plotted against time and the counting rate was obtained by the slope of the curve. Background and reference materials were also counted and treated in the same way. Results showed that ^{226}Ra varied from 45 to 116 Bq kg^{-1}; ^{228}Ra, from 80 to 165 Bq kg^{-1}; ^{40}K, from 155 to 1 187 Bq kg^{-1} and ^{137}Cs varied from 0.3 to 7 Bq kg^{-1}. The methodology applied for determining low levels of ^{137}Cs in sediment proved to be efficient and reproducible.

Keywords: Salesópolis, natural radionuclides, artificial radionuclides, reservoir.

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1. INTRODUCTION

Environmental radioactivity occurs mainly due to the presence of natural radionuclides of the uranium and thorium series and ^{40}K in soil, rocks, water and air along with artificial nuclides also spread in the biosphere after nuclear accidents and nuclear weapon tests [1]. Although in the South Hemisphere nuclear tests have been fewer in number than that in the North, artificial radionuclides can also be found spread at ground level [2, 3]. The distribution of the radionuclides once released in the environment to the different compartments depends on their different geochemical behaviors, as they are subjected to a number of natural processes such as dissolution, adsorption, complexation, and oxidation/reduction.

The natural and man made radionuclides may eventually be accumulated in sediments due to processes like weathering and terrestrial minerals and rocks recycling, rainfall and other depositional phenomena such as gravitational settling and precipitation [4]. Sediments, in turn, can act as sink for these elements or release them to the surrounding water depending on the physical-chemical conditions. This paper aims to evaluate the distribution of the radionuclides belonging to the U and Th series, ^{40}K and the artificial ^{137}Cs in a sediment core of the Salesópolis reservoir by gamma spectrometry.

Salesópolis is located in the São Paulo city Metropolitan Region (SPMR). The Usina Parque Rio Tietê (Salesópolis) reservoir belongs to the Alto do Tietê system for the capture, storage and treatment of water for the SPMR. Therefore, the quality of the water, as well as of sediments of this dam is of great importance.

2. MATERIALS AND METHODS

A sediment core named T1B with 42 cm depth was collected in the Salesópolis dam in 2.014 with a manual core sampler under a water column of approximately one and a half meters. In the laboratory, the core was sliced each 3 cm totalizing 14 samples. Each sample was then dried in a ventilated oven at a temperature of 45 °C and passed in a sieve of 2 mm aperture. After that, approximately 25 g of samples were weighted in plastic containers of 5 cm diameter and 1.5 cm of

height, sealed and let to wait at least 30 days for the radioactive equilibrium between ^{226}Ra and its daughters to be reached.

Gamma activity was measured in a EG&G Ortec spectrometer with relative efficiency of 60%, and resolution of 1.16 and 2.25 keV for the 122 and 1 332 keV photopeaks of ^{57}Co and ^{60}Co, respectively. In order to diminish the background (BG) contribution the detector is shielded with a sequential layer of lead and copper combined protection.

The activity concentration of ^{226}Ra was determined by the mean activity measurement of its daughter's photopeaks in 351 keV (^{214}Pb), 609 and 1.120 keV (^{214}Bi). The activity concentration of ^{228}Ra was determined by the gamma transitions in 338 keV, 911 keV (^{228}Ac), 238 keV (^{212}Pb) and 727 keV (^{212}Bi). Potassium-40 and ^{137}Cs were determined directly by means of their gamma transition photopeaks in 1.460 keV and 661 keV, respectively.

The efficiency curve determination was performed by using the reference materials IAEA-RGU (uranium ore diluted with floated silica powder), IAEA-RGTh (thorium ore diluted with floated silica powder), IAEA-K (high purity potassium sulphate) and IAEA-375 (radionuclides and trace elements in soil) in the same geometry as the samples for the same gamma transitions photopeaks to be determined. All these reference material have activity sufficiently high to give a statistical error of less than 1% in the considered counting time. No self-attenuation correction was applied since the energy of all determined nuclides was higher than 200 keV.

For the data acquisition, the processing software was programmed to save the counting files from 100 000 s to 160 000 s in intervals of 10 000 s. A background measurement was also done intercalating each two samples counting. This BG counts was subtracted from the samples counting [5].

For each sample and respective BG a plot of gross counts per time was generated to obtain the curve slope. Figures 1 and 2 exemplify the procedure for ^{137}Cs determination and that was then repeated for each radionuclide to be determined.

Calculation of the activity concentration was done according to the formula presented in equation 1.

$$A_i = \frac{C_i - C_{bgi}}{m_a x \varepsilon_i} \quad (1)$$

where:

A_i is the activity concentration related to the photopeak i (Bq kg^{-1})

C_i is the slope of the curve obtained for the photopeak i in the sample
C_{bgi} is the slope of the curve obtained for the photopeak i in the background
m_a is the sample mass (kg)
ε_i is the counting efficiency for the photopeak of interest.

Figure 1: *Plot of the background counts per time (in seconds) in the 661 keV region of interest*

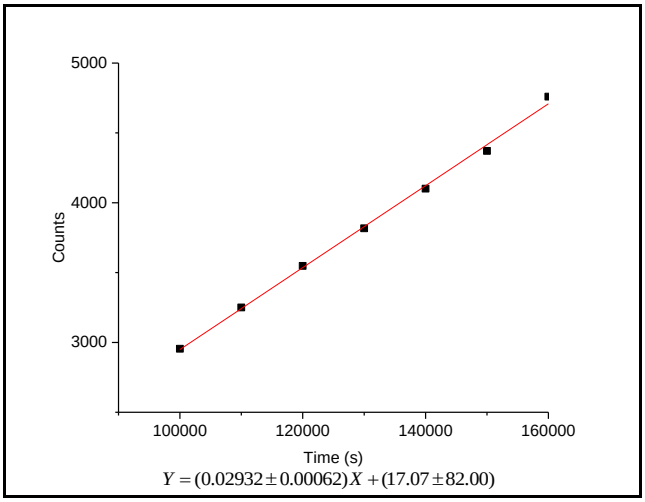
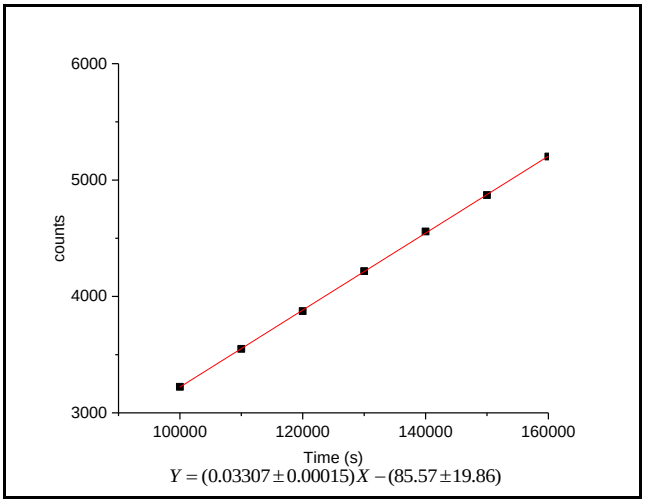


Figure 2: *Plot of the counts per time (in seconds) in the 661 keV region of interest for a given sample*



The uncertainties were determined by error propagation, with the uncertainties of C*i* and C_{bgi} being that obtained from the slope of the adjusted curve for each photopeak considered.

3. RESULTS AND DISCUSSION

Table 1 shows the activity concentration measured in the reference material IAEA-414, IAEA-SD-N-2 and IAEA-375 and it is possible to see the good agreement between the measured values and the certified ones.

Table1: Results obtained for the reference materials IAEA-327, IAEA-414 and IAEA-SD-N-2, in Bq kg^{-1} (recommended values in parenthesis).

	Reference material	Obtained value	recommended value
^{226}Ra	IAEA-327	43 ± 8	38.7 ± 5.2
^{228}Ra	IAEA-327	40 ± 7	38.7 ± 4.7
^{40}K	IAEA-327	641 ± 41	621 ± 52
	IAEA-414	406 ± 26	481 ± 16.5
	IAEA-SD-N-2	218 ± 14	220 ± 16
^{137}Cs	IAEA-327	28 ± 7	24.9 ± 1.8
	IAEA-414	5 ± 1	5.18 ± 0.1
	IAEA-SD-N-2	0.9 ± 0.2	0.8 ± 0.5

3.1. Natural radionuclides

Table 2 presents the values of the determined radionuclides in the samples obtained in the analyzed core. It is possible to see that radium isotopes are in higher concentrations than that reported for the global soil average [6] of 32 Bq kg^{-1} for ^{226}Ra and 45 Bq kg^{-1} for ^{228}Ra (considered in equilibrium with ^{232}Th). For ^{40}K, only the first 12 cm are below the global average of 420 Bq kg^{-1} [6] and the activity concentrations tend to rise with the depth.

Figure 3 shows that the radium isotopes have almost the same behavior with depth, with the lowest values observed between 9 and 12 cm depth. Higher activity concentrations were also observed for both nuclides in the base of the core. Nevertheless, ^{228}Ra tend to be enriched over ^{226}Ra in the top layers with the enrichment being less pronounced towards the core base as depicted by

the $^{228}Ra/^{226}Ra$ ratio showed in Figure 4 probably indicating a change in the sediment source and mineralogy.

Few data are available on the activity concentrations of natural radionuclides in Brazilian reservoirs. Concentrations lower than those observed in this study, around 20 Bq kg^{-1}, were determined for ^{226}Ra in the Apipucos Reservoir, Recife, Brazil [7]. In sediments of Cubatão river, ^{226}Ra activity concentration varied in the range of 28 to 80 Bq kg^{-1} [8] and the activity concentrations of ^{226}Ra and ^{228}Ra were in the range of 26 to 37 Bq kg^{-1} and 59 to 74 Bq kg^{-1}, respectively [9]. Considering aquatic environments with saline water, levels of ^{238}U (^{226}Ra), ^{232}Th and ^{40}K of 21, 28 and 426 Bq kg^{-1} were determined [10] in the shelf and upper slope of Southeast Brazil. Also levels of ^{226}Ra in the range of 4.2 to 28.5 were determined in the Cananéia- Iguape system [11]. Mean activity concentrations of 24 Bq kg^{-1} for ^{226}Ra, 44 Bq kg^{-1} for ^{232}Th and 678 Bq kg^{-1} for ^{40}K were found in the Piraquara de Fora Inlet located in Ribeira Bay within the Ilha Grande Bay in Angra dos Reis, Rio de Janeiro State [3]. It can be seen that the values obtained in this study for the Salesópolis reservoir are generally higher that those reported in literature and that sediments of salty waters generally presents lower activity concentrations.

Table 2: Activity concentrations, in Bq kg^{-1}, of measured radionuclides in the sediment core samples.

Depth (cm)	^{226}Ra	^{228}Ra	^{40}K	^{137}Cs
0-3	74±13	126±31	300±19	5±1
3-6	67±12	132±33	155±10	2±1
6-9	72±13	105±26	272±17	7±3
9-12	45±8	80±20	179±11	0.3±0.1
12-15	67±12	113±20	680±43	3±1
15-18	79±14	133±33	807±52	4±2
18-21	65±11	108±27	856±55	3±1
21-24	72±13	116±29	843±54	3±1
24-27	70±12	103±26	830±53	4±2
27-30	79±11	122±30	978±63	4±2
30-33	83±14	130±32	1 026±66	4±2
33-36	75±13	122±30	940±60	4±2
36-39	89±15	124±31	950±61	5±2
39-42	116±20	165±41	1 187±76	4±2

Table 3 shows a compilation of activity concentrations for radium isotopes, ^{40}K and ^{137}Cs, found in sediments of reservoirs located in North Hemisphere. For the natural radionuclides it can be seen

that the levels are closer to those observed for the global average [6] and present a mean narrower those observed for the Brazilian reservoir and the results obtained in this work.

Higher activity concentrations for natural radionuclides may be related to geological and geochemical conditions such as the rock and soil basement and leaching by an intense rainfall system.

Figure 3: *Distribution of ^{226}Ra and ^{228}Ra with the depth in the analyzed core*

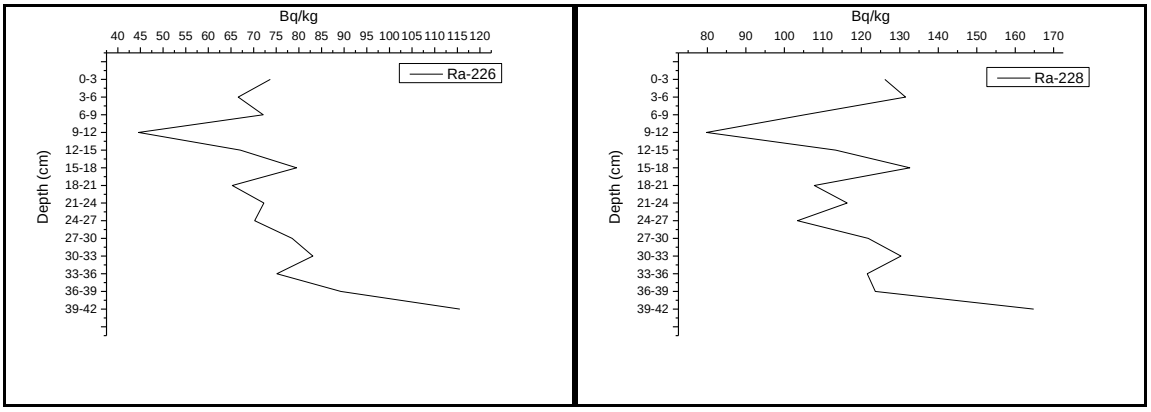
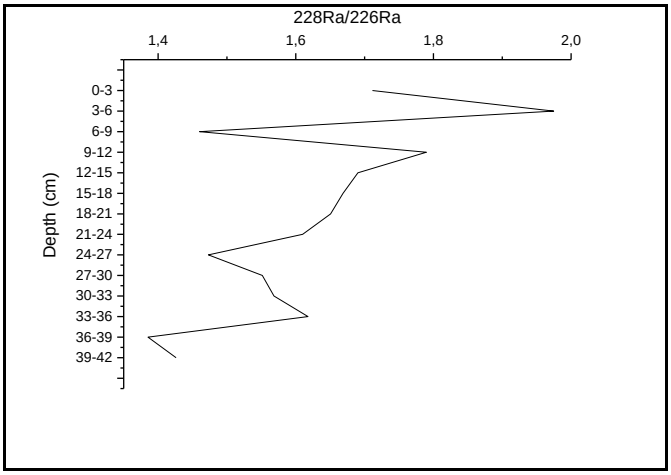


Figure 4: *Distribution of $^{228}Ra/^{226}Ra$ isotopic ratio with depth*



3.2 Artificial radionuclide ^{137}Cs

The distribution of ^{137}Cs with depth is shown is Figure 5. The lowest activity concentration was observed at 9 – 12 cm depth and the highest was determined in the range of 6 to 9 cm, although it is

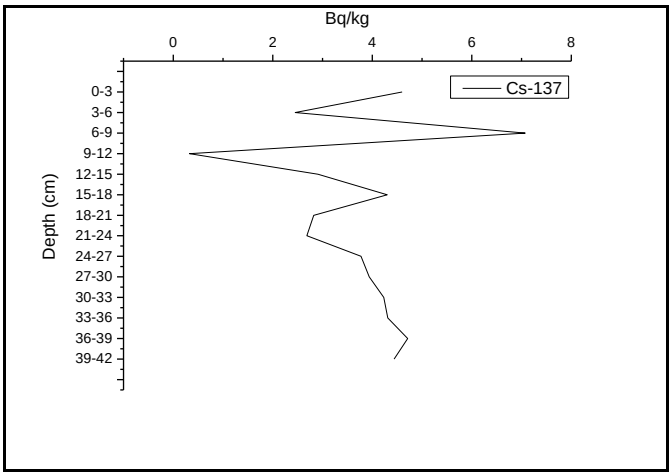
unlikely that this peak corresponds to the maximum fallout of Cs. The observed variation in the top layers may be related to the grain size and organic matter distribution [18].

The most probable peak of ^{137}Cs activity concentration corresponding to the fallout of 1963-1964 can be observed in the depth of 36 – 39 cm. As for the natural radionuclides, few activity concentrations were reported for ^{137}Cs in reservoir sediments.

Table 3: Activity concentrations, in Bk kq^{-1}, of natural and artificial radionuclides in sediment samples of reservoirs form selected locations in the North Hemisphere.

	^{226}Ra	^{228}Ra	^{40}K	^{137}Cs	Reference
Spain	58 - 94	59 - 116	535 - 823	1.7 - 60.2	[12]
India	4 - 26	5.5 - 19.9	250 - 558	3.6 - 54.5	[13]
Tunisia	23 - 29.8	40 - 52		0.9 - 3.9	[14]
Serbia	8 - 29	2.7 - 45		0.8 - 140	[15]
China	31.2 - 37.5	56.5 - 79.8	179.4 - 214.9		[16]
Poland				89 - 865	[17]
Poland				30 - 243	[18]
Turkey				1 - 101	[19]

Figure 5: *Distribution of ^{137}Cs with the depth in the analyzed core*



Considering Brazilian sediment of other aquatic environments, levels of this radionuclide in the range of 0.2 to 6.1 Bq kg^{-1} were found in the Cananéia-Iguape system [11]; activity concentration of 0.5 ± 0.2 Bq kg^{-1} for ^{137}Cs was determined in the Piraquara de Fora Inlet [3]; values around 3 Bq kg^{-1} were found in the sediments of Guajará Bay, in Pará State [2] and a maximum of 4.7 Bq kg^{-1}

was reported [20] for the Brazilian cost. The values obtained in this study are in good agreement with those observed in the references cited above, and all the highest values found in Brazilian sediments were much lower than those found in the North Hemisphere, as shown in Table 3. Closer values to the ones reported here were found in a South African reservoir with activity concentrations around 10 Bq kg^{-1} [21].

4. CONCLUSIONS

The methodology applied for determining low levels of ^{137}Cs in sediment proved to be efficient and reproducible. The levels of natural radionuclides found in the sediment core of Salesópolis dam are higher than the world average. These activities can be related to the local geochemistry associated with intense rainfall. Results showed a tendency of raising values in the core base possibly indicating a variation in the mineralogical features as shown by the decrease in the $^{228}Ra/^{226}Ra$ ratio.

The activity concentrations of ^{137}Cs present a great variation in the upper layers probably related to the granulometry distribution and organic matter content. The values found are in accordance with the ones observed for other Brazilian aquatic environments. Nevertheless, more studies are necessary to better understand the observed variation as well as the hole of local conditions in the observed values.

5. ACKNOWLEDGMENT

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