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Natural radionuclides in soil profiles and sediment cores from Jundiaí reservoir, state of Sao Paulo.

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ABSTRACT

The activity concentration of natural radionuclides in soils and sediments is dependent on many factors, such as the rock parental material, pedogenic and weathering processes, physical and chemical properties of the environment, anthropogenic sources, among other aspects. There are few studies about the levels of natural radionuclides in reservoirs in both, international and national, literature. The objective of this paper was to evaluate the activity concentrations of ^{238}U and ^{232}Th by Instrumental Neutron Activation Analysis and ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th and ^{40}K by gamma spectrometry in two soil profiles and three sediment cores collected in the catchment area of Jundiai reservoir, located in the state of São Paulo, Brazil. Principal component analysis was applied to verify the correlation of the activity concentrations of the natural radionuclides with physical and chemical properties of soil and sediment samples. The mean activity concentrations of the radionuclides in the soil profiles 1 and 2 were, respectively: ^{238}U – 37(1) and 32(1) Bq.kg^{-1}; ^{232}Th – 91(1) and 60(1) Bq.kg^{-1}; ^{226}Ra – 66(1) and 51(1) Bq.kg^{-1}; ^{210}Pb – 35(1) and 37(1) Bq.kg^{-1}; ^{228}Ra – 34(1) and 27(1) Bq.kg^{-1}; ^{228}Th – 78(1) and 58(1) Bq.kg^{-1}; ^{40}K – 96(2) and 171(7) Bq.kg^{-1}. For the three sediment cores analyzed, the average activities concentrations of the radionuclides were, respectively: ^{238}U – 64(0.5), 47(0.1) and 44(0.2) Bq.kg^{-1}; ^{232}Th – 122(2), 100(1) and 64(1) Bq.kg^{-1}; ^{226}Ra – 74(2), 71(1) and 45(1) Bq.kg^{-1}; ^{210}Pb – 70(3), 56(2) and 55(2) Bq.kg^{-1}; ^{228}Ra – 53(1), 41(1) and 33(1) Bq.kg^{-1}; ^{228}Th – 100(2), 92(1) and 63(2); ^{40}K – 316(5), 237(1) and 136(2) Bq.kg^{-1}.

Keywords: Natural radionuclides, Jundiaí reservoir, gamma spectrometry.

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1. INTRODUCTION

Ionizing radiation is characterized by electromagnetic waves and electrically charged particles which ionize other atoms. Natural radionuclides are formed by sources of ionizing radiation [1] and these elements can be founded in all types of ecosystems of the environment. Human and environmental exposure to natural radiation is an important subject for public health prevention, geologic characterization, and radioecology studies [2]. The naturally occurring radiation materials are part of the Earth's rocks composition. These rocks undergo weathering and erosion processes and carried out radionuclides and other trace elements for soil composition. These elements can be transported, by water or wind, to a sedimentation watershed, where occurs the deposition in the bottom of a lake, ocean, rivers, or reservoirs [3] and absorbed by biota, causing concern about the possible negative biological effects that the radiation can trigger in animals, plants or humans [4].

Natural radionuclides can be found in different minerals; in igneous and metamorphic rocks, these elements are presented mostly in minerals like apatite, titanite and zirconite, and also in uraninite, monazite, allanite, pyrochlore and thorite, but these minerals are rare [5]. Sedimentary rocks are composed of shale, sandstone, detrital rocks and others, and those with high amounts of clay fractions have higher concentrations of uranium and thorium [6].

The use of fertilizers in soils can increase the activity concentrations of radionuclides from the ^{238}U and ^{232}Th decay series, as well as the concentration of the natural radionuclide ^{40}K [1]. Hence, the major source of human exposure to radiation is the natural source, mostly due to the radionuclides ^{226}Ra, ^{210}Pb, ^{228}Th, ^{228}Ra present in both series, also ^{40}K. Another source of natural radiation is cosmogenic radionuclides; however, its contribution to human exposure is lower [2]. External exposure is due to the gamma-ray emission and internally exposure occurs through inhalation or ingestion, including water consumption [2].

Reservoirs are constructed by river segmentation and its impacts are phenomena to study [7], including the fact that the natural radionuclides transportation and deposition are influenced by these modifications in the environment [4]. This paper had a purpose to evaluate the activity concentrations of ^{238}U and ^{232}Th by Instrumental Neutron Activation Analysis - INAA, and ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th and ^{40}K by Gamma-ray Spectrometry in soils profiles and sediment cores from

Jundiai reservoir. The behavior of the natural radionuclides in the soil profiles and the sediment cores was studied using statistic correlation of soil and sediment physical-chemical properties, grain size, organic matter content and pH, with the radionuclide vertical distribution.

2. MATERIALS AND METHODS

2.1. Study area and sampling

The Jundiai reservoir is located at the southeast region of the state of Sao Paulo in Mogi das Cruzes county (23°40'S and 46°10'W); it is formed by river Biritiba-Mirim and Tietê waters, has a drainage area of 111 km^2 and a flooded area about to 17 km^2. The local climate is classified as a temperate ocean climate in the Koppen system (Cfb), with annual mean rainfall and temperature of 1582 mm and 17.6°C, respectively [8]. The Upper Tietê water basin is responsible to supply millions of people in the Sao Paulo Metropolitan area. This area is characterized by intense agriculture, industrial activities [9], and the concentration of many pollutants that can affect the natural radionuclide concentrations. Soil samples were collected vertically in two profiles located near to the Jundiai reservoir shore and sampled each 10 cm. Sediment samples were collected in three points in the margin of the reservoir with 70-100 cm of the water column with a Polyvinyl Chloride (PVC) core sampler; two of these sampling points of the sediment cores represent coordinates from the respective soil profile. A third sediment core was collected away from the location of the other soil profiles. The sampling information is presented in Table 1.

Table 1: Sampling points, geographical coordinates type of matrix, sampling date, depth of the profiles and cores, soil taxonomy [11], and sample designation.

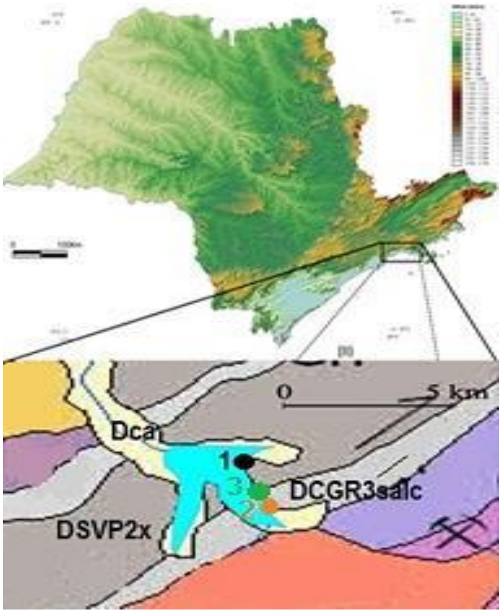
Point	**Geographical coordinates**	**Matrix**	**Sampling date**	**Depth**	**Soil Taxonomy**	**Designation**
1	23°37'25"S 46°10'19"W	Soil/Sediment	27.03.2018	0.9m/0.29m	Dystrophic Red-Yellow Ultisoil	JUSO1/JUSE1
2	23°38'35"S 46°10'0"W	Soil/Sediment	27.03.2018	1.4m/0.28m	Dystrophic Red-Yellow Ultisoil	JUSO2/JUSE2
3	23°38'36"S 46°10'5"W	Sediment	27.03.2018	0.4m	-	JUSE3

The sampling points were chosen according to the main lithology around the reservoir. Figure 1 presents the Jundiai reservoir and the lithology map [10] with the three sampling points. The geological domain around the reservoir is characterized by metamorphic Proterozoic volcano-sedimentary sequences represented by silty-clay metasediments (DSVP2x) and intensely deformed granitic complexes of the sub alkaline series (DGCR3salc). The flooded area of the reservoir is represented by unconsolidated Cenozoic sediments deposited in a recent alluvial plain (Dca).

The soil samples were packed in low-density polyethylene (LDPE) bags and taken to *Laboratório de Radiometria Ambiental* (LRA-IPEN/USP) for the analysis; at the laboratory, they were air-dried and passed through a 10 mesh (2.00 mm) sieve. Afterward, the samples were homogenized and an aliquot of one quarter was reserved.

The sediment samples were obtained after opening the collected sediment cores, and the sediment was sampled according to the morphology; the samples were dried in an oven at 50°C.

Figure 1: *Jundiaí reservoir lithology map and sampling points.*



Source: adapted from PERROTA *et al.*, 2005.

2.2. Physical-chemical parameters of soil and sediment

Soil and sediment grain size analysis were performed by sieving 5 g of the samples and the particles were classified as sand (0.06 mm to 2 mm), silt (2 µm to 0.06 mm) and clay (<2 µm). Soil and sediment textures were classified according to the USDA-NRCS [11] ternary diagram for soil texture.

Soil and sediment organic matter contents – OC - were estimated by the loss-on-ignition method. The samples were dried at high temperatures in a muffle furnace and the organic carbon volatilizes as CO_2. The content of organic matter was evaluated by gravimetric calculations based on mass loss; the samples were weighed before and after the heating at 500°C. This method provides only an estimate of the total organic carbon in samples; other compounds, such as carbonates and sulfates can be lost on the process [12].

The potential of hydrogen ion activity (pH) of soil samples was measured using a pH meter with 0.02 uncertain. The measurement was performed using potassium chloride (1 mol. L^{-1}) and calcium chloride (0.01 mol. L^{-1}) with a soil/extractor proportion of 1:5. The pH measured in calcium chloride solution represents the active acidity of soil; the solubility of elements and geochemical speciation are followed by active acidity. The exchangeable soil acidity was measured using the solution of potassium chloride, and this compartment is correlated with the amount of exchangeable Al^{3+} adsorbed in clay minerals and humic substances present in the soil composition. The delta pH (ΔpH) was measured according to the expression (1) and represents the electrical charge present in soil colloid [13].

$$\Delta pH = pH_{KCl} - pH_{CaCl_2} \tag{1}$$

Where:

$\Delta pH < 0$; surface area of colloid is negative electrically charged (cation adsorption).

$\Delta pH > 0$; surface area of colloid is positive electrically charged (anion adsorption).

$\Delta pH = 0$; zero point of charge (ZPC).

2.3. INAA, Gamma-ray Spectrometry and statistical analysis

The activity concentrations of ^{238}U and ^{232}Th were determined by Instrumental Neutron Activation Analysis - INAA. For the analysis, the aliquots of soil and sediment were mashed, using mortar and pestle, in 115 mesh (0.125 mm), and approximately 200 mg of samples and the reference materials Lake Sediment – SL3 from IAEA, Inorganic Marine Sediment – 2702 and

Montana Soil II, both from NIST (National Institute of Standards and Technology), were irradiated in a thermal neutron flux of 1012 n.cm^{-2}. s^{-1} at the IPEN Research Reactor IEA-R1 for a 6 h period. The measurement process was performed by Gamma-ray Spectrometry, using a coaxial HPGe detector, ORTEC, and associated electronics. The analysis of the spectrum was done with *ORTEC INTERWINNER^{TM} 6.0* [X1] gamma-ray software to identify and discriminate the gamma-ray peaks.

The activity concentrations of the radionuclides ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th, and ^{40}K were determined by Gamma-ray Spectrometry. Approximately 120g of the soil samples and 15g of the sediment were measured in a coaxial Be-layer HPGe detector with 25% relative efficiency, 2.09 keV resolution at 1.33 MeV and associated electronic devices, with a live counting time of 150 000s (Soil samples were packed in polyethylene bottles, while sediments were placed in Petri dishes). The spectra were acquired by multichannel analyzer Ethernim and, for the analysis, WinnerGamma software was used [14]. The associated uncertainty for one sigma confidence ranged from 10% to 40% from the obtained results. The detector was calibrated using IAEA 300 Standard Reference Material.

For the statistical analysis, the Principal Component Analysis - PCA, which is a multivariate statistical method based on a linear distribution reduction of the variances in two principal components, was used. It is advantageous for the simultaneous analysis of several variables and this technique shows which parameter has more statistical significance in the analysis; the x component represents more significance of variance that the y component [15]. This method was employed to establish statistical correlations between the activity concentrations of the radionuclides and the physical-chemical parameters of soil and sediment samples.

3. RESULTS AND DISCUSSION

3.1 Soil

The soil physical-chemical parameters are presented in Table 2. The mean content of clay, silt and sand fractions for the first and second profiles were 45%, 5%, 44% and 28%, 13%, 55%, respectively. The first profile, JUSO1, presented a higher amount of organic matter (OC) in comparison with JUSO2; the mean contents for these profiles were 13% and 9%, respectively. The potential of hydrogen ion analysis showed positive values of ΔpH at most depths analyzed, except in surface samples, which presented negative values of ΔpH. The mean active acidity (pH in $CaCl_2$) exhibited similar values in the two profiles analyzed, with an average of 3.86 and 3.87, respectively, whereas the mean exchangeable acidity (pH in KCl) values were higher (4.10 and 4.13).

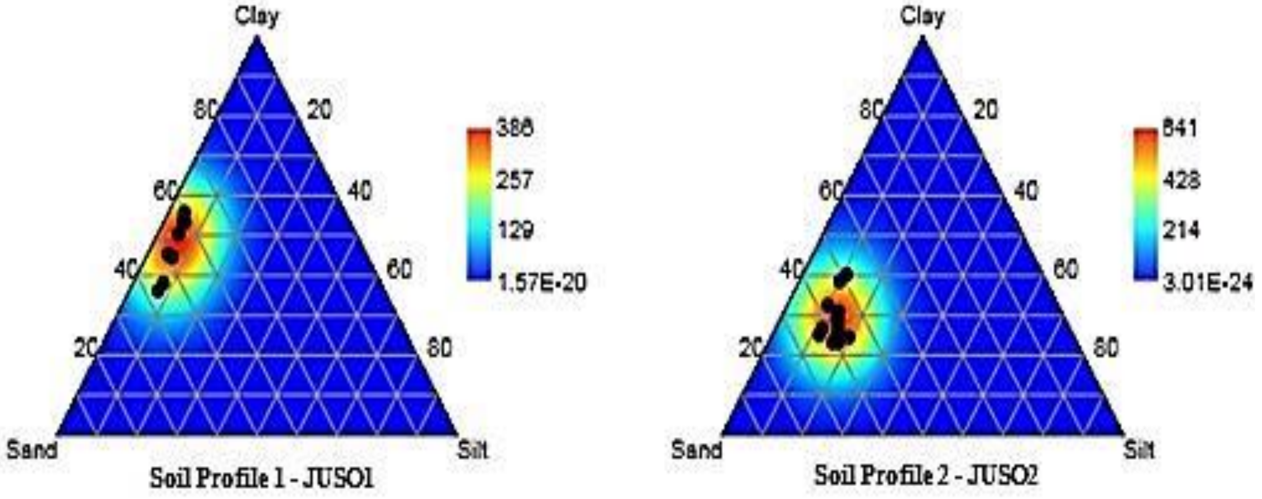
Table 2: Grain size analysis, potential of hydrogen-ion activity, organic matter contents of the soil profiles analyzed.

JUSO1	**Grain size (%)**			**Potential of hydrogen ion Activity**[*]			**Organic matter Content (%)**
cm	**Clay**	**Sand**	**Silt**	**pH KCl**	**pH $CaCl_2$**	**ΔpH**	
0	33	52	7	3.67	3.69	-0.02	12.5
10	42	46	5	3.88	3.72	+0.16	25.3
20	42	46	6	3.91	3.75	+0.16	11.1
30	35	50	7	3.94	3.72	+0.22	11.6
40	43	47	6	3.94	3.80	+0.14	10.8
50	48	43	5	4.09	3.79	+0.30	11.5
60	49	38	5	4.10	3.82	+0.28	11.4
70	50	39	4	3.96	3.86	+0.10	11.7
80	50	39	5	4.01	3.80	+0.21	9.9
90	53	39	4	4.02	3.76	+0.26	10.4
JUSO2							
0	24	61	11	3.29	3.31	-0.02	16.7
10	26	51	13	3.40	3.45	-0.05	12.8
20	27	54	14	3.63	3.57	+0.06	10.8
30	25	58	11	3.74	3.66	+0.08	8.2
40	23	53	18	3.79	3.70	+0.09	8.6
50	22	58	15	3.81	3.74	+0.07	7.5
60	24	58	16	3.89	3.78	+0.11	7.5
70	27	58	16	3.90	3.74	+.016	6.8
80	22	57	17	3.93	3.76	+0.17	7.3
90	28	51	12	3.97	3.81	+0.16	7.4
100	29	55	13	3.98	3.81	+0.17	7.0
110	30	55	13	4.02	3.84	+0.18	7.4
120	32	56	10	4.06	3.74	+0.32	6.9
130	37	50	10	4.07	3.80	+0.27	7.1
140	39	47	11	4.13	3.87	+0.26	6.7

[*] For the potential of hydrogen-ion activity analysis was used a table pH meter with 0.05 uncertain. All samples were weighted using an analytical balance with two decimal places.

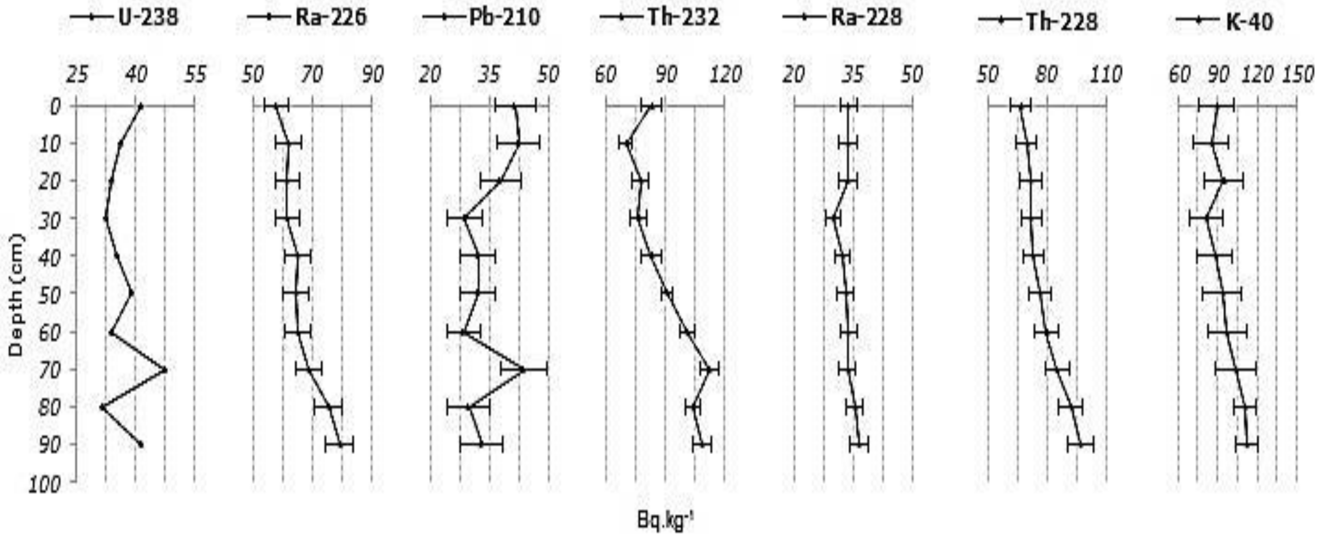
According to USDA (1999), soil textures were classified as sandy clay and sandy clay loam [11] for JUSO1 and JUSO2, respectively, as shown in Figure 2.

Figure 2: Ternary diagram of the soil profiles textures according to USDA-NRCS classification.



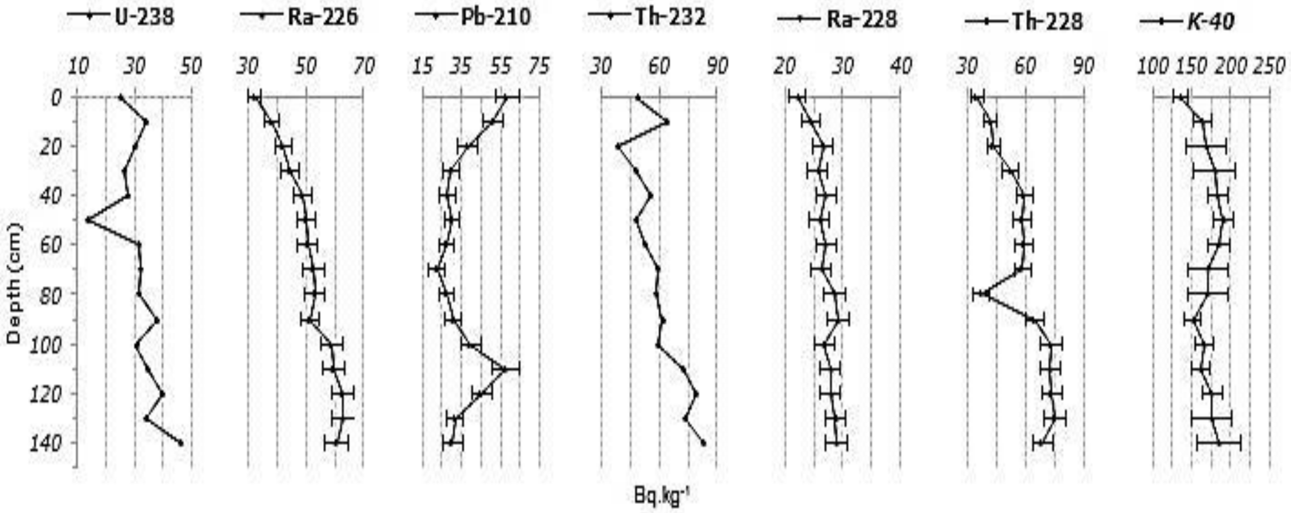
Figures 3 and 4 present the results obtained for the activity concentrations of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{228}Ra, ^{228}Th, and ^{40}K, in Bq.kg^{-1} with associated expanded uncertainties (k=2), in the function of the depth (cm), for JUSO1 and JUSO2.

Figure 3: Activity concentrations, in Bq.kg^{-1}, of the natural radionuclides in soil profile 1 – JUSO1.



*The expanded uncertainties values for ^{238}U were lower than 5% in all depths analyzed.

Figure 4: Activity concentrations, in Bq.kg^{-1}, of the natural radionuclides in soil profile 2 – JUSO2.



*The expanded uncertainties values for ^{238}U and ^{232}Th were lower than 5% in all depths analyzed.

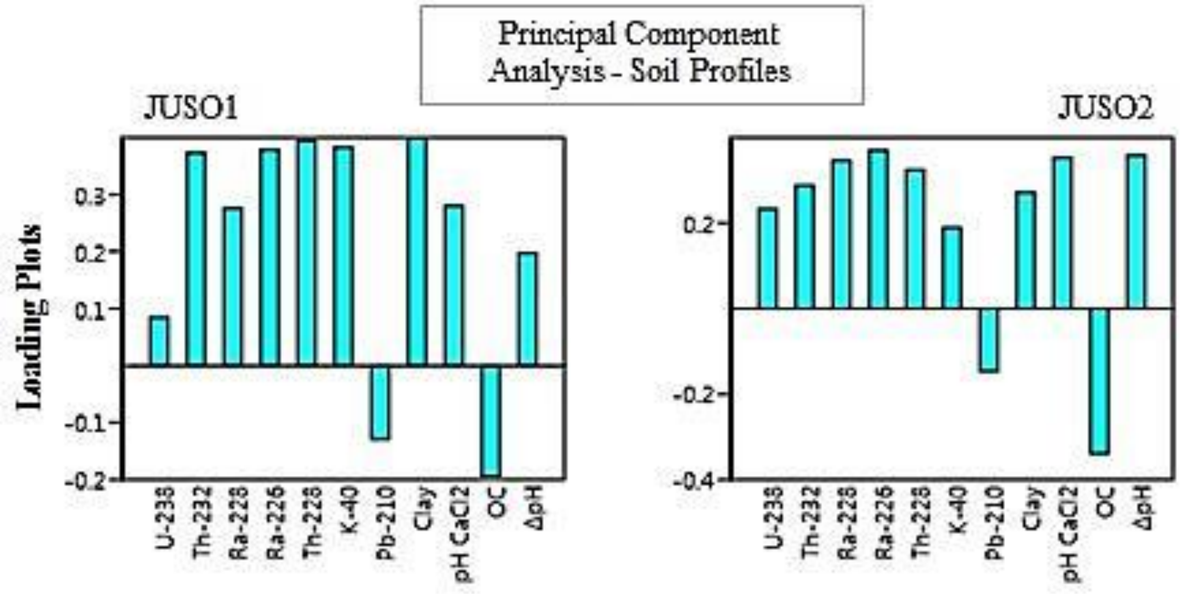
In both profiles, the activity concentration of the radionuclide ^{232}Th presented higher values when compared to UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) values [2]. The mean activity concentrations for ^{232}Th in JUSO1 and JUSO2 were 90 ± 1 Bq.kg^{-1} and 60 ± 1 Bq.kg^{-1}, respectively. According to UNSCEAR, the mean activity concentration of ^{232}Th in global soils is 30 Bq.kg^{-1}. The results obtained in this work cannot be considered an anomaly, because the values from UNSCEAR were obtained by various types of soils and different types of lithology around the Earth.

The radionuclides ^{226}Ra, ^{228}Ra, and ^{40}K presented levels in the range of the activity concentration for these radionuclides in global soils. The mean activity concentration of ^{238}U presented slightly higher values than the UNSCEAR for JUSO1. The mean activity concentration of ^{238}U obtained in this profile was 37 ± 0.3 Bq.kg^{-1}, and the mean level in global soils is 35 Bq.kg^{-1}.

The mean activity concentrations of ^{210}Pb obtained in JUSO1 and JUSO2 were 35 ± 1 Bq.kg^{-1} and 37 ± 1 Bq.kg^{-1}, respectively; in both profiles, the levels of this radionuclide were higher in the surface, probably due to ^{222}Rn decay and subsequent deposition of ^{210}Pb.

Figure 5 presents the results obtained in PCA between the activity concentrations of the natural radionuclides and the soil physical-chemical parameters.

Figure 5: Principal Component Analysis (PCA) for the soil profiles.



[*] Positive values of loading plots are associated with the coordinate that embrace the more significant variance of the principal component analysis.

In both soil profiles, the results of PCA showed that the parameters representing greater significance for the availability of radionuclides in the soil were the percentage of clay fraction and the pH of the active acidity (pH in $CaCl_2$). Data from the literature [16] show that the rise of soil alkalinity provides an increase in soil cation exchange capacity (CEC), which could explain this strong association. The chemical behavior of the radionuclides is similar to metals behavior, and metals, normally, forms cationic compounds in the environment. The organic matter showed good association with ^{210}Pb, however, this correlation represents less significance over the total variance. For JUSO1, the ΔpH parameter presented a negative association with the activity concentration of ^{210}Pb. In the surface of the profiles, the values of ΔpH were negative, and this indicates that the surface area of soil colloid is negative electrically charged, which favors the cation adsorption.

The radium isotopes, ^{228}Ra and ^{226}Ra, presented a strong association with the clay fraction and the thorium isotopes (^{232}Th and ^{228}Th) showed an inverse correlation with the content of organic matter. Thorium becomes readily more soluble when it complexes with the organic chelates [17], present in high amount in the soil surface; hence, this radionuclide could leach to deeper horizons of the soil profile, where the contents of organic matter are lower.

3.2 Sediment

The results of the physical-chemical parameters of the sediment cores are presented in Table 3 and the texture classifications in Figure 6. The first core - JUSE1- presented higher content of organic matter than the others.

The mean content of organic matter in JUSE1, JUSE2 and JUSE3 was 6%, 5%, and 4%, respectively. The mean contents of clay, silt and sand fractions for JUSE1 were 23%, 16%, and 57%; for JUSE2 were 13%, 12%, and 73%; for JUSE3 were 13%, 12%, and 73%, respectively. These values of grain size analysis classified the sediment cores as sandy clay loam texture, JUSE1; sandy clay loam texture, JUSE2 and sandy loam, JUSE3.

Figure 6: Ternary diagram of the sediment cores textures according to USDA-NRCS classification.

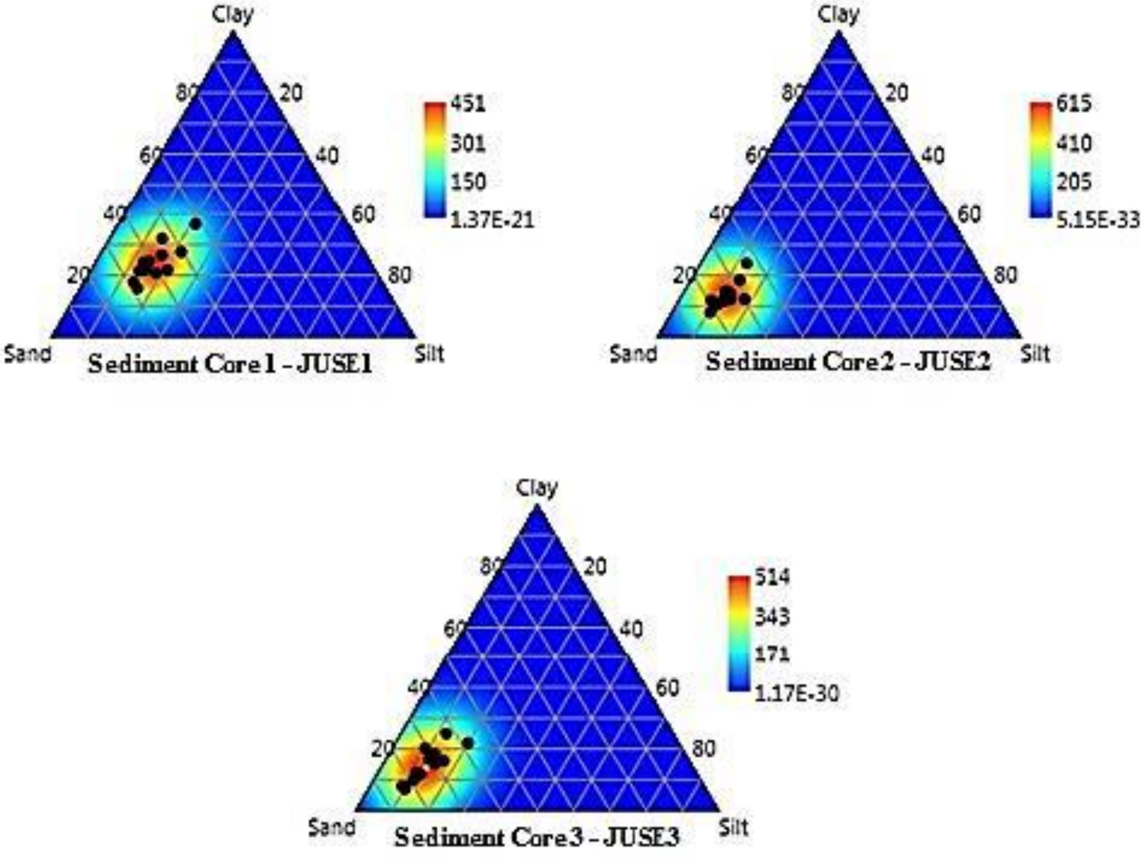


Table 3: Grain size analysis and organic matter contents of the sediment cores analyzed.

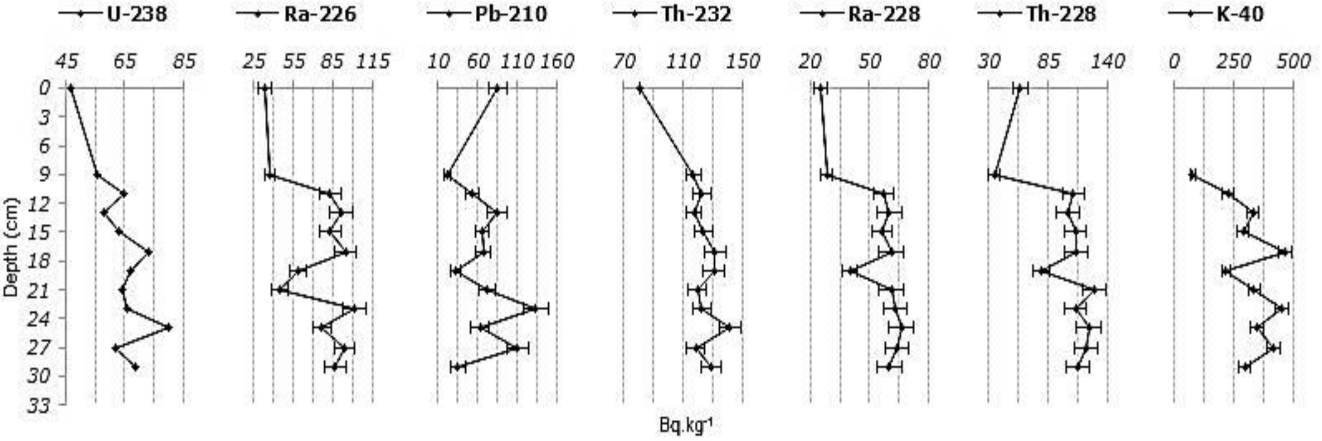
JUSE1	**Grain size (%)**			**Organic matter Content (%)**
cm	**Clay**	**Sand**	**Silt**	
0	16	68	16	25.8
9	24	60	14	4.8
11	25	53	16	4.3
13	34	39	20	4.7
15	31	52	14	3.2
17	24	62	13	4.4
19	17	67	13	4.7
21	21	62	13	4.8
23	21	61	15	4.2
25	27	49	21	3.7
27	21	56	20	3.0
29	20	60	18	3.6

JUSE2	**Grain size (%)**			**Organic matter Content (%)**
cm	**Clay**	**Sand**	**Silt**	
0	12	77	9	11.1
4	8	79	10	7.7
6	10	75	11	6.2
8	14	70	11	4.7
10	15	73	12	4.0
12	18	68	13	3.8
14	14	74	12	3.6
16	24	63	13	3.5
18	12	67	17	3.6
20	11	72	11	3.6
22	14	72	13	4.0
24	12	72	14	3.5
26	12	75	13	3.6
28	12	75	13	4.2

JUSE3	**Grain size (%)**			**Organic matter Content (%)**
cm	**Clay**	**Sand**	**Silt**	
0	11	78	11	9.3
4	12	77	12	7.2
7	10	80	11	3.3
10	7	82	10	2.1
13	8	84	9	1.7
16	8	82	9	3.0
19	24	60	12	6.5
22	12	74	10	9.0
25	17	72	12	5.8
28	20	70	9	4.7
31	19	71	13	1.6
34	16	74	15	1.4
37	17	71	17	2.7
40	22	58	20	4.7

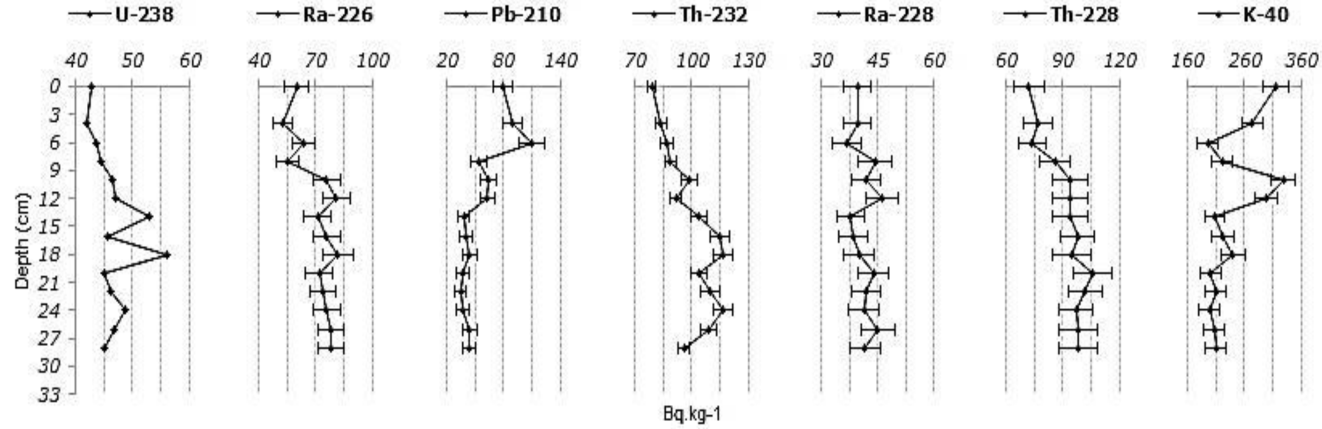
Figures 7, 8 and 9 present the results obtained for the activity concentrations of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{228}Ra, ^{228}Th, and ^{40}K, in Bq.kg^{-1} with associated expanded uncertainties (k=2), in the function of the depth (cm), for JUSE1, JUSE2 and JUSE3.

Figure 7: Activity concentrations, in Bq.kg^{-1}, of the natural radionuclides in sediment core JUSE1.



*The expanded uncertainties values for ^{238}U were lower than 5% in all depths analyzed.

Figure 8: Activity concentrations, in Bq.kg^{-1}, of the natural radionuclides in sediment core JUSE2.

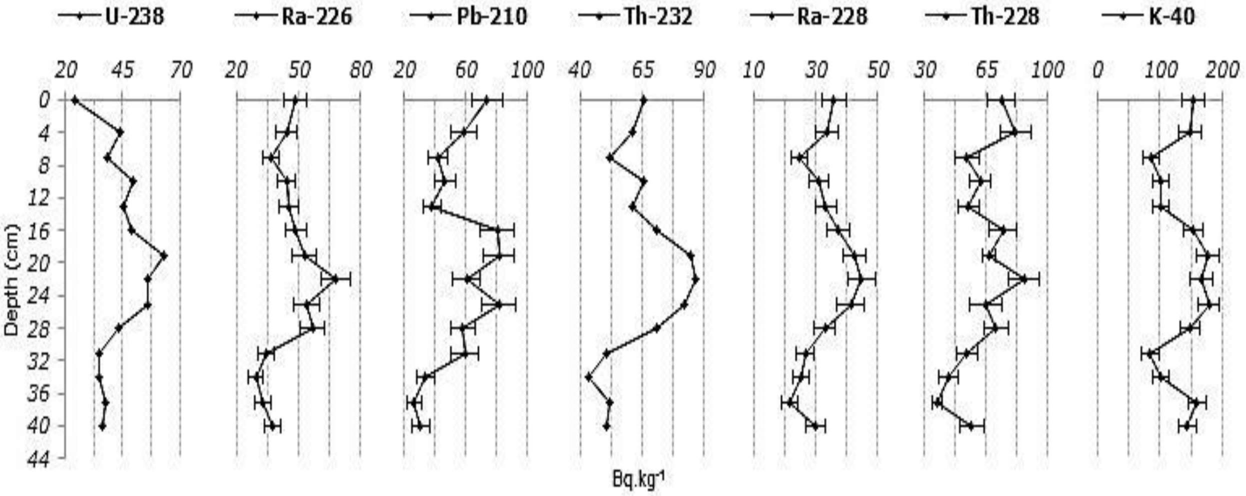


*The expanded uncertainties values for ^{238}U were lower than 5% in all depths analyzed.

The mean activity concentrations of the radionuclides ^{238}U and ^{232}Th were higher than the UNSCEAR values in the three sediment cores. The mean activity concentrations of ^{238}U in JUSE1, JUSE2 and JUSE3 were 64 ± 0.5 Bq.kg^{-1}, 47 ± 0.1 Bq.kg^{-1}, and 44 ± 0.2 Bq.kg^{-1}, respectively. For

^{232}Th, the results of mean activity concentrations obtained in JUSE1, JUSE2 and JUSE3 were 122 ± 2 Bq.kg^{-1}, 100 ± 1 Bq.kg^{-1}, and 64 ± 1 Bq.kg^{-1}, respectively.

Figure 9: Activity concentrations, in Bq.kg^{-1}, of the natural radionuclides in sediment core JUSE3.



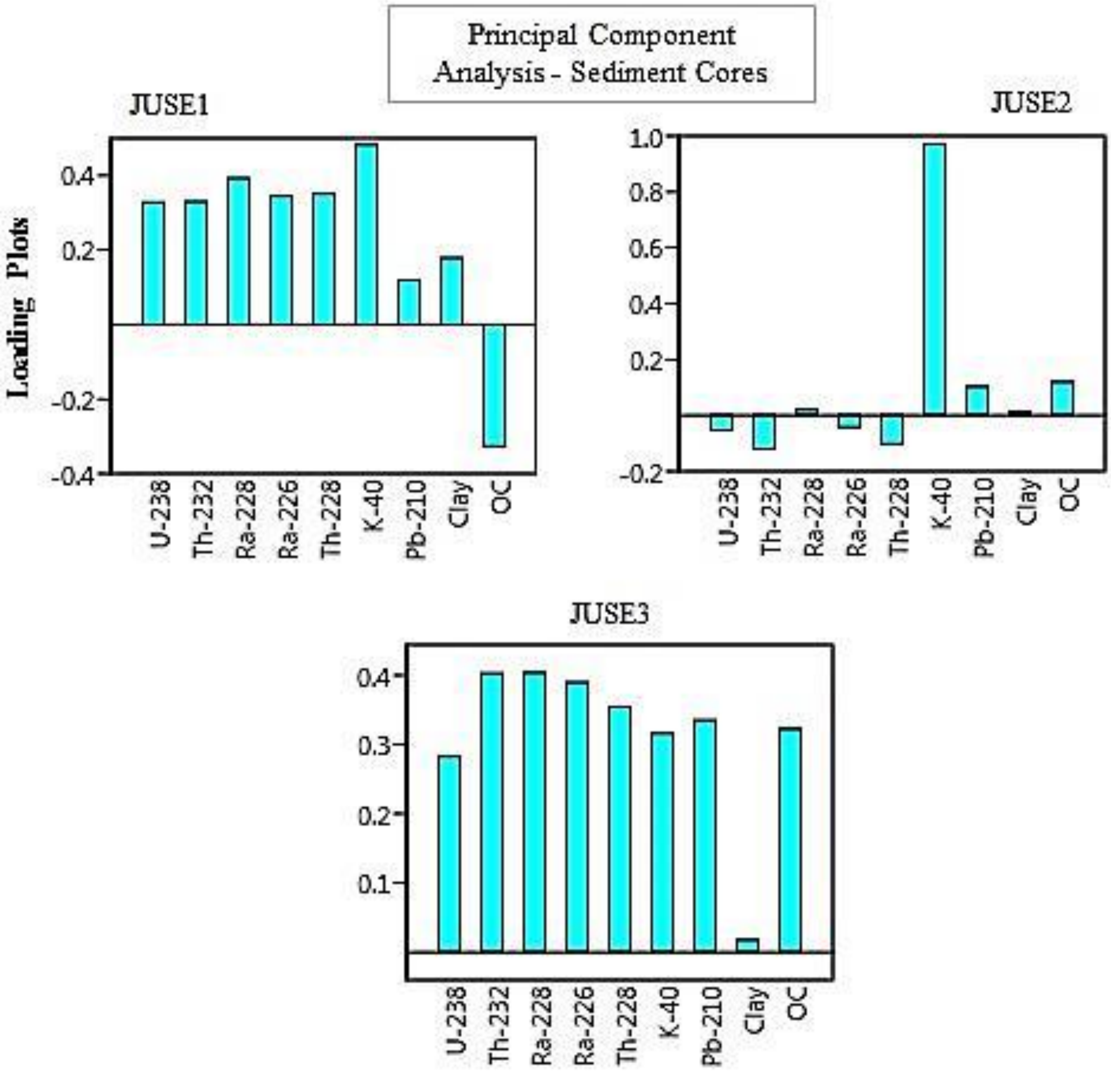
*The expanded uncertainties values for ^{238}U and ^{232}Th were lower than 5% in all depths analyzed.

Another radionuclide that showed higher levels when compared to UNSCEAR values was ^{226}Ra; its values were higher in JUSE1 and JUSE2, and the mean activity concentrations obtained were 74 ± 2 Bq.kg^{-1} and 71 ± 1 Bq.kg^{-1}, respectively. According to UNSCEAR, the variation of activity concentration for ^{226}Ra in global soils is between 17 Bq.kg^{-1} to 60 Bq.kg^{-1}; these were unexpected results due to the high amount of sand in the sediment cores and because the radium retention should be reduced in these conditions.

The activity concentrations of ^{210}Pb presented high variations in JUSE1 and JUSE3 and this variation could be associated with the variations of dry periods in the region of Sao Paulo after the construction of the Jundiai reservoir. The dry period has a strong influence in the flooded area of the reservoir and when marginal sediment gets exposed to air, the radon gas (^{222}Rn) emanates and decays to a several short half-life radionuclides until ^{210}Pb that is carried out by the rainwater. Figure 10 presents the results obtained in PCA between the activity concentrations of the natural radionuclides and the sediment physical-chemical parameters for JUSE1, JUSE2 and JUSE3.

In general, the organic matter contents presented more statistical significance over the clay fraction for the availability of the natural radionuclides in the sediment cores; however, in JUSE1 and JUSE2, this association was negative. In these two cores, the amount of clay showed an insignificant association with the activity concentrations of the natural radionuclides. Other factors in the environment, such as physical-chemical parameters of the reservoir water, could be inducing the distribution of the radionuclides in the sediments. Therefore, more detailed research is needed for a complete understanding of these radionuclide behaviors in this specific environment.

Figure 5: Principal Component Analysis (PCA) for the sediment cores.



4. CONCLUSION

The present work evaluated the activity concentrations of the natural radionuclides from ^{238}U and ^{232}Th series and ^{40}K in soil profiles and sediment cores collected in the Jundiaí reservoir. Some natural radionuclides presented the results in higher activity concentration than UNSCEAR values: In the soil profiles, the activity concentration of ^{232}Th presented higher levels when compared to the levels in global soils, and in the sediment cores, the activity concentrations of ^{232}Th and ^{238}U were also higher, however, only the activity concentrations of radium isotopes demand more detailed research and investigation. These values were higher than expected and the high contents of sand in sediment cores should reduce the radium retention, despite both radium isotopes are soluble.

The principal component analysis performed to verify the behavior of the natural radionuclides showed good associations of activity concentrations with soil pH and clay fraction in the soil profiles. In the sediment cores, other parameters need to be evaluated to explain the availability of the natural radionuclides.

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