

NATURAL RADIONUCLIDES FROM U-238 AND TH-232 SERIES AND INORGANIC CHEMICAL CHARACTERIZATION OF SOIL PROFILES AND SEDIMENT CORES OF THE TAIACUPEBA RESERVOIR, SÃO PAULO, BRAZIL

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ABSTRACT

Taiacupeba reservoir, located in the state of São Paulo, Brazil, belongs to Producer System of Alto Tietê (Sistema Produtor Alto Tietê) and it is responsible for water supply for about 3.1million of people. The water quality of a reservoir is very important, but this is reduced by the increase of environmental degradation of the soil around the reservoir and its different uses. The study of soil profiles and sediment cores is an important tool for understanding the geophysical and geochemical aspects of an aquatic ecosystem. The objective of this work was to present the natural radionuclides ²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²³²Th, ²²⁸Th, ²²⁸Ra and ⁴⁰K activity concentrations and also the inorganic chemical characterization of four soil profiles and four sediment cores collected in the area of influence area of Taiacupeba reservoir. The analytical techniques, gamma spectrometry and instrumental neutron activation analysis were used in the determination. In the soil profiles the highest activity concentrations were obtained for the radionuclides ⁴⁰K and ²²⁸Th and the lowest for ²¹⁰Pb; in the sediment cores the highest activity concentrations were obtained for the radionuclide ²¹⁰Pb and the lowest for ²²⁶Ra and ²²⁸Ra. For the inorganic chemical characterization the highest values obtained were for Na, As and Sb; in a sediment core a very high concentration was obtained for the element Zn indicating a probable accumulation of this element inside the reservoir; enrichment factor was used to evaluate a possible anthropic contamination in the soil and sediment at the margins of Taiacupeba reservoir.

1. INTRODUCTION

Taiacupeba reservoir, located in the state of São Paulo, on the border of the municipalities of Mogi das Cruzes and Suzano, is part of the Production System Alto Tietê - SPAT, along with Ponte Nova, Jundiá, Biritiba and Paraitinga reservoirs. The waters of these reservoirs are delivered through channels and tunnels, being collected in Taiacupeba reservoir by ETA (water treatment station) of Sabesp for treatment and distribution to approximately 3.1 million people of east zone of the city of São Paulo and of the municipalities of Arujá, Itaquaquecetuba, Poá, Ferraz de Vasconcelos, Suzano, Mauá, Mogi das Cruzes, part of Santo André and the Pimenta and Bonsucesso neighborhoods of the city of Guarulhos [1-3].

The evaluation of water quality of a reservoir has become of great importance in the last decades due to the increase of the environmental degradation of the surrounding soils, due to the construction of houses in its margins and to the diverse usages of the reservoir, like irrigation, fishing, industrial and population supply, as well as obtaining electricity, recreation, among others. Water quality can be also influenced by many factors, including the river basins that form the reservoir, the climate, the volume of precipitation and vegetation.

The soil around the reservoir and human activities upstream also influence the quality of the water, as a reservoir behaves as a collector of all activities that may occur in its surroundings. In the soil, can be found natural radionuclides from ^{238}U and ^{232}Th decay series, as well as other radionuclide, for example ^{40}K [4-6], which are one of the major sources of natural radiation exposure to which the human being is exposed [7]. These natural radionuclides, as well as the chemical elements present in the rocks of which the soil originated, is transferred to other compartments in the environment, through the chemical and physical weathering being incorporated to the water and accumulating later in the sediment.

The sediment is composed of particles of different sizes, shapes and chemical composition, and it is deposited, according to their textural properties in the bottom of the aquatic environments. Sediment also contains precipitated materials of chemical and biological processes that occur in these ecosystems [8-9]; in addition, it constitutes a sector of great potential for the accumulation of trace elements and when these trace elements accumulate at high levels can indicate a pollution of the ecosystem [10-13].

The study of soil and sediment profiles is an important tool for understanding the geochemical and geophysical aspects of aquatic ecosystems [14], since the analysis of these compartments is of great importance in the study of human exposure to natural radiation and also in the study of environmental pollution.

The objective of this work was to determine the natural radionuclide concentrations of ^{238}U , ^{226}Ra , ^{210}Pb , ^{232}Th , ^{228}Th , ^{228}Ra and ^{40}K and also the inorganic chemical characterization (determining the elements As, Ba, Br, Ca, Ce, Co, Cr, Cs, Fe, Hf, K, La, Lu, Na, Nd, Rb, Sb, Sc, Ta, Tb, Th, U, Yb and Zn) in four soil profiles and four sediment cores collected in the influence area of Taiacupeba reservoir using the analytical techniques gamma spectrometry and instrumental neutron activation analysis (INAA), respectively. The enrichment factor was used to evaluate a possible anthropic contamination in the soil and sediment at the margins of Taiacupeba reservoir.

2. METHODOLOGY

2.1 Sampling and pre-treatment of the samples

Four soil profiles, TASSO 01, TASSO 02, TASSO 03 and TASSO 04, and four sediment cores, TASSO 01, TASSO 02, TASSO 03 and TASSO 04, were collected in the influence area of Taiacupeba reservoir, in points that best represent the local lithology: point 1 ($23^{\circ} 36' 33.36''$ S and $46^{\circ} 16' 8.94''$ W), point 2 ($23^{\circ} 35' 35.09''$ S and $46^{\circ} 17' 28.68''$ W), point 3 ($23^{\circ} 35' 13.95''$ S and $46^{\circ} 17' 20.13''$ W) and point 4 ($23^{\circ} 35' 50.20''$ S and $46^{\circ} 16' 48.33''$ W), respectively, Fig. 1. The explanation of the lithology codes shown in Fig. 1 (2) can be found in Perrota et. al [15]. The soil profiles were collected near the margin of the reservoir and the

sediment cores inside the reservoir at a water column depth of approximately 80 cm.

Soil profiles were collected in trenches up to 2 meters deep and sampled every 5 cm. In the laboratory the samples were dried at room temperature; the sediment samples were collected with a manual PVC sampler sliced every 2 cm or according to their textural characteristics, and dried in an oven at 50 °C. The length of the sediment cores ranged from 42 cm to 61 cm. After drying, soil and sediment samples were sieved in a 2 mm mesh and packed in polyethylene bottles for further analysis.

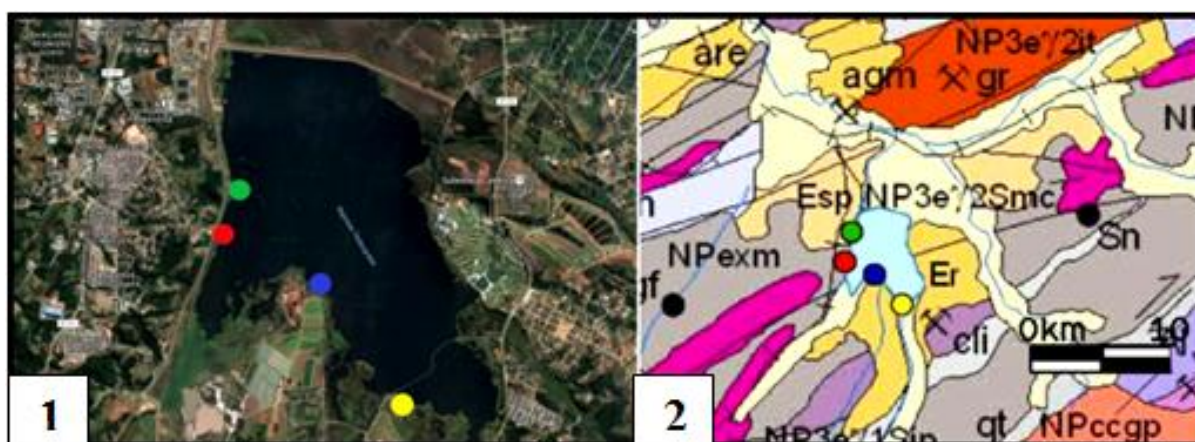


Figure 1: Satellite image of Taiacupeba reservoir (1), lithology (2) [15], location of the soil and sediment collection points, 01 - yellow, 02 - red, 03 - green and 04 - blue

2.2. Grain Size Analysis

The grain size analysis was performed wet by sieving 5 g of soil or sediment sample with MiliQ - water in sieves that allow the separation of the fractions, sand, silt and clay [16].

2.3 Gamma spectrometry

The gamma spectrometry was used to determine the concentrations of the natural radionuclides ^{226}Ra , ^{210}Pb , from ^{238}U decay series and ^{228}Th and ^{228}Ra from ^{232}Th decay series, and also ^{40}K . Approximately 100 g of soil and 15 g of sediment were used for the analysis, which were homogenized and sealed in polyethylene bottles and Petri dishes, respectively. Samples were stored for a period of 30 days to ensure equilibrium between the ^{226}Ra and their short half-life descendants, and measured over a period of 150.000 seconds. A hyperpure germanium detector (HPGe) XTRa 2520 model, CANBERRA, and associated electronics were used for measurement. Spectra were analyzed using the program InterWinner-WinnerGamma 6.0 ORTEC, and the concentration of each radionuclide in the sample was calculated from the net area. [16].

2.4 Instrumental neutron activation analysis – INAA

The instrumental neutron activation analysis - INAA was used to determine the radionuclides ^{238}U and ^{232}Th activity concentrations and As, Ba, Br, Ce, Co, Cr, Cs, Eu, Fe, Hf, La, Lu, Na, Nd, Rb, Sb, Sc, Sm, Ta, Tb, Th, U, Yb, and Zn concentrations in both soil profiles and sediment cores. In the sediment analysis were used the reference materials, Lake Sediment, SL1, of the International Atomic Energy Agency – IAEA and Montana II from NIST; for the

soil analysis, in addition to the reference materials cited for the sediment, Basalto Ribeirão Preto, BRP1, of UNICAMP was also used.

For analysis, approximately 200 mg of each sample and reference material were weighed separately into previously cleaned polyethylene bags. These were irradiated for a period of 6 h, under a thermal neutron flux of 10^{12} n cm⁻²s⁻¹, at Research Reactor IEA-R1 from IPEN. Concentration measurements were performed on a Hyperpure Germanium detector (HPGe - INTERTECHNIQUE) with associated electronic and InterWinner-WinnerGamma 6.0 - ORTEC was used for the spectra analysis. The elements concentration in the samples were obtained, in mg.kg⁻¹, by comparing the peak areas of the samples and the activated reference materials together with the samples [16].

2.5 Enrichment Factor – EF

The evaluation of the Enrichment Factor (EF) was used to verify if the natural elements present in the soil and sediments studied are enriched, indicating possible anthropic contamination. In the present work values from the Upper Continental Crust (UCC) [17] were used as background and the element Sc was used as normalizer element. The enrichment factor was calculated by the expression (1) [16].

$$FE = \frac{(C_i/C_n) \text{ sample}}{(C_i/C_n) \text{ background}} \quad (1)$$

Where:

- EF: Enrichment factor;
- C_i: concentration of the interest element;
- C_n: concentration of the normalizer element.

The parameters used for this assessment follow the range described by Sutherland [18]. In this paper will be presented only the results for moderate enrichment, very high enrichment and very severe enrichment.

3. RESULTS AND DISCUSSION

3.1 Soil

3.1.1 Gamma spectrometry

Fig. 2 shows the natural radionuclides ²²⁶Ra, ²¹⁰Pb, ²²⁸Ra, ²²⁸Th and ⁴⁰K activity concentrations obtained by gamma spectrometry, as well as the radionuclides ²³⁸U and ²³²Th activities concentrations obtained by INAA, for the soil profiles TASO 01, TASO 02, TASO 03 and TASO 04. It can be observed that in all profiles the highest activity concentrations were obtained for the radionuclide ⁴⁰K, mainly in TASO 03 profile, followed by ²²⁸Th and ²³²Th; the lowest activity concentrations were obtained for ²¹⁰Pb.

3.1.2 Instrumental neutrons activation analysis (INAA)

The chemical element concentrations in soil profiles TASO 01, TASO 02, TASO 03 and TASO 04 are shown in Fig. 3, 4, 5 and 6, respectively.

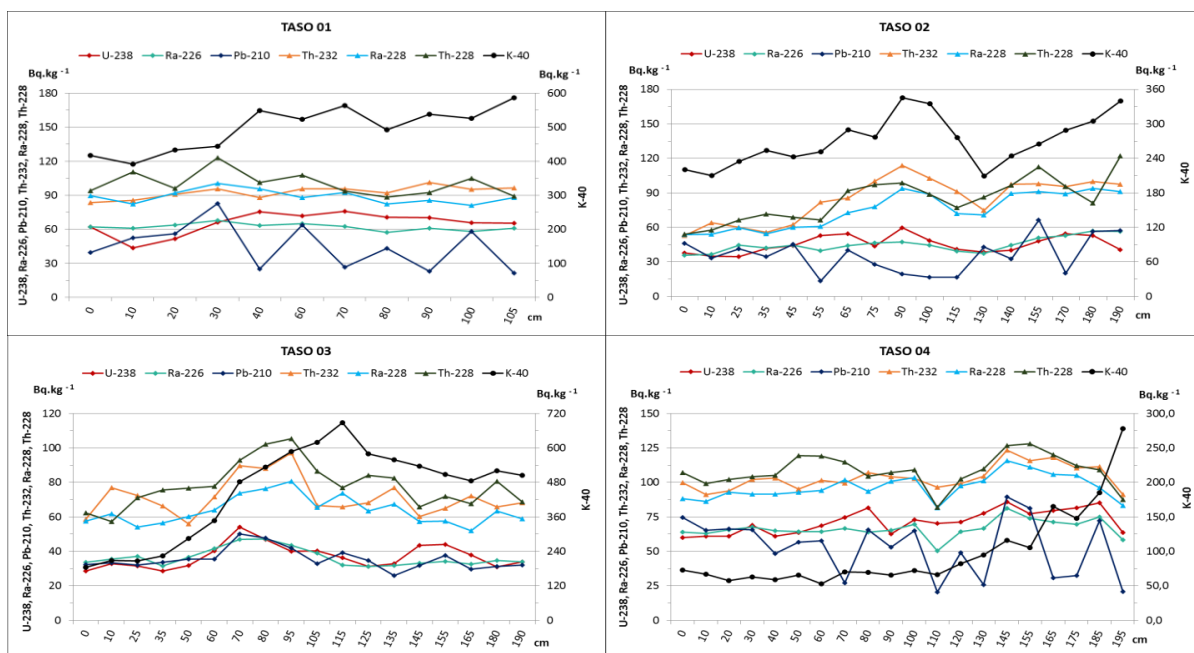


Figure 2: Activity concentration of ^{238}U , ^{226}Ra , ^{210}Pb , ^{232}Th , ^{228}Ra , ^{228}Th and ^{40}K for TASO 01, TASO 02, TASO 03 and TASO 04 soil profiles.

TASO 01 profile, Fig. 3, presented an increase in the concentration of the elements Eu, Rb, Cs, Sm and Nd with the depth and constant concentrations for the other elements; the elements Ba, Zn and Co showed a significant increase between the depths 10 cm and 30 cm.

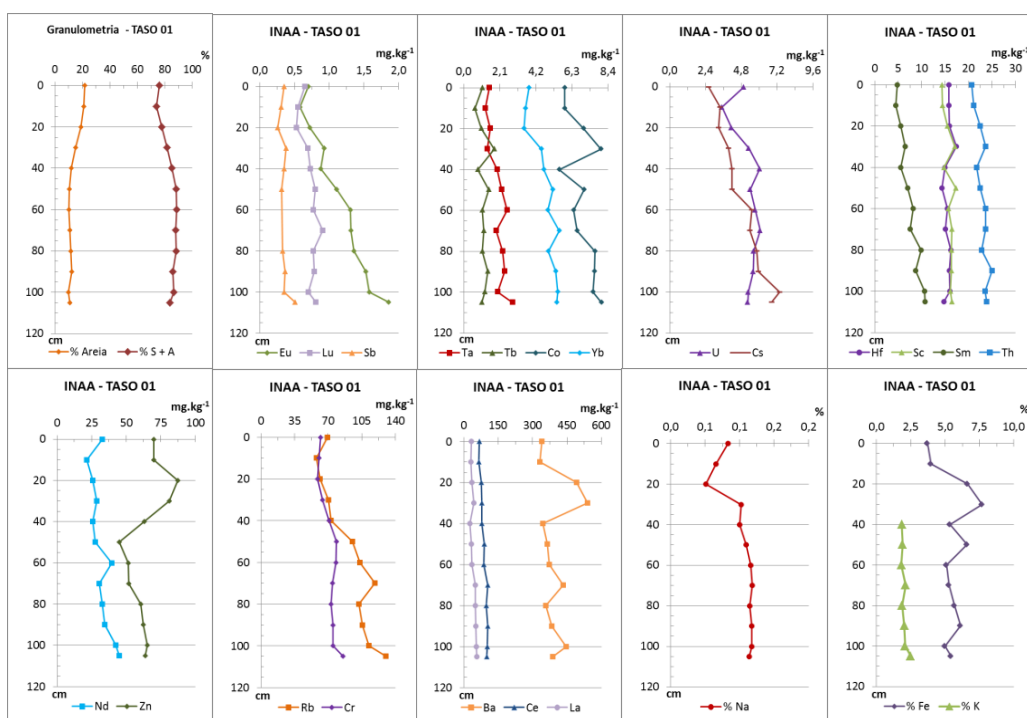


Figure 3: Concentrations of the elements obtained by INAA, in TASO 01 profile.

TASO 02, Fig. 4, presented an increase in the concentrations of the elements Eu and La with depth, and variations in the concentrations of the elements Cs, Sc, Th, Rb and Cr, as

presented by the thin fraction of the profile. The concentration of the other chemical elements in the profile remained constant.

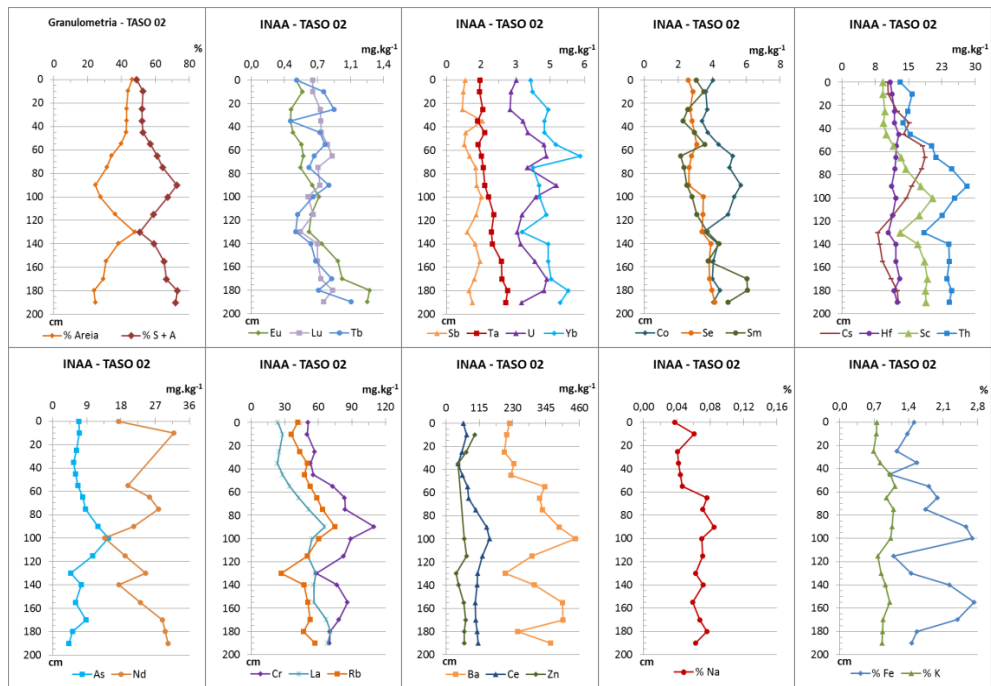


Figure 4: Concentrations of the elements obtained by INAA, in TASO 02 profile.

TASO 03, Fig. 5, presented increase in the concentrations of Lu, Ta, U, Sm, Sb and Th elements between depths 60 cm and 100 cm and similar values for the beginning and end of the profile. The concentrations of the other elements increased with depth, and higher in the middle of the profile according to the fine fraction.

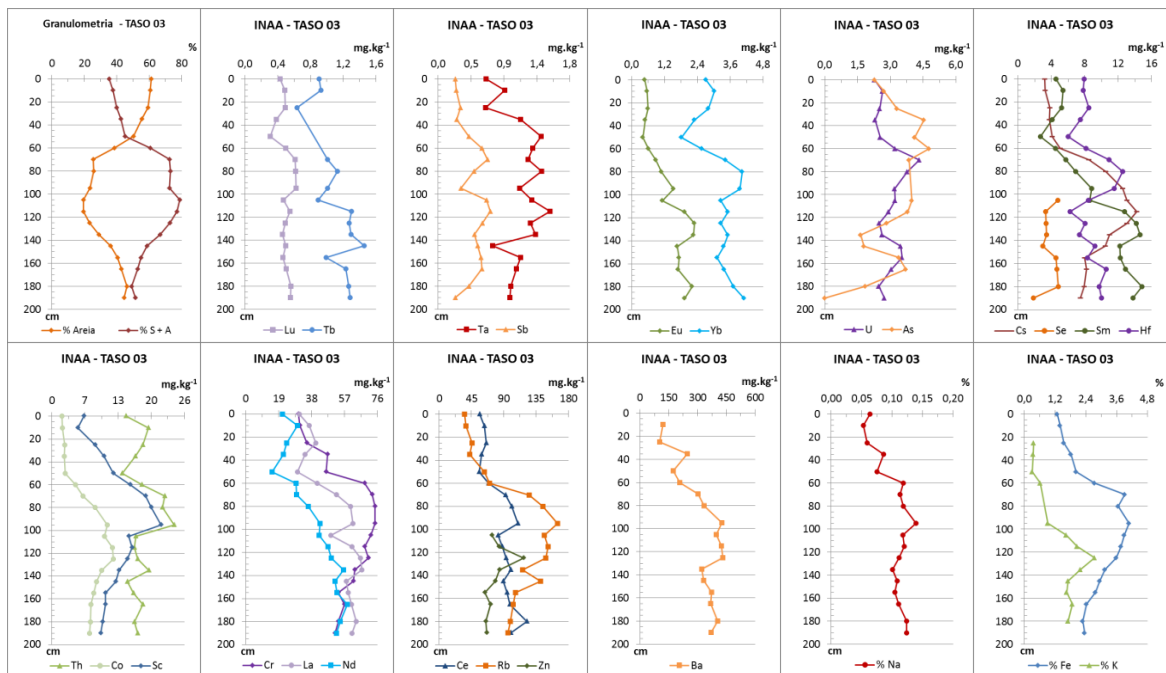


Figure 5: Concentrations of the elements obtained by INAA, in TASO 03 profile.

TASO 04, Fig. 6, presented practically constant concentrations for the majority of the chemical elements, a tendency also observed for the fine fraction of the soil. An increase in the Na element concentration and reduction in As element concentration at the end of the profile was also observed. The elements Tb and Se showed increase and sharp reduction between 60 cm and 120 cm depth.

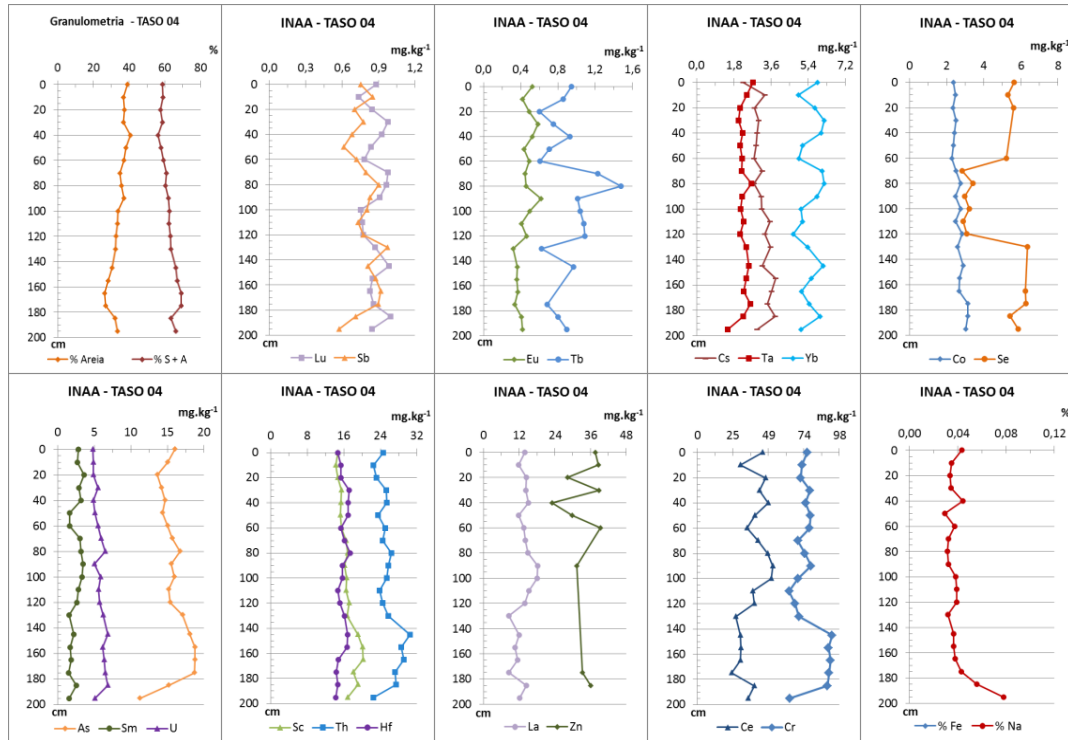


Figure 6: Concentrations of the elements obtained by INAA, in TASO 04 profile.

3.1.3 Enrichment Factor – EF

The obtained results of EF for the soil profiles TASO 01, TASO 02, TASO 03 and TASO 04 are presented in Fig.7.

TASO 01 showed very high enrichment for the element Na and moderate enrichment for the element Hf. TASO 02 showed very high enrichment for the element Na and for the elements As and Sb in some profile slices and moderate enrichment for the elements Cs, Yb, Hf and Lu, with higher values at the beginning of the profile.

TASO 03 presented very high enrichment for the element Na and moderate enrichment for the elements As, Cs, Eu, Sm, La, Sb, U, Ce, Hf, Nd, Th, Yb, Lu and Tb with higher values of FE at the top and bottom of the profile. TASO 04 showed very high enrichment for the elements As and Na at the beginning and end of the profile, and moderate enrichment for the elements Sb, Hf and Lu.

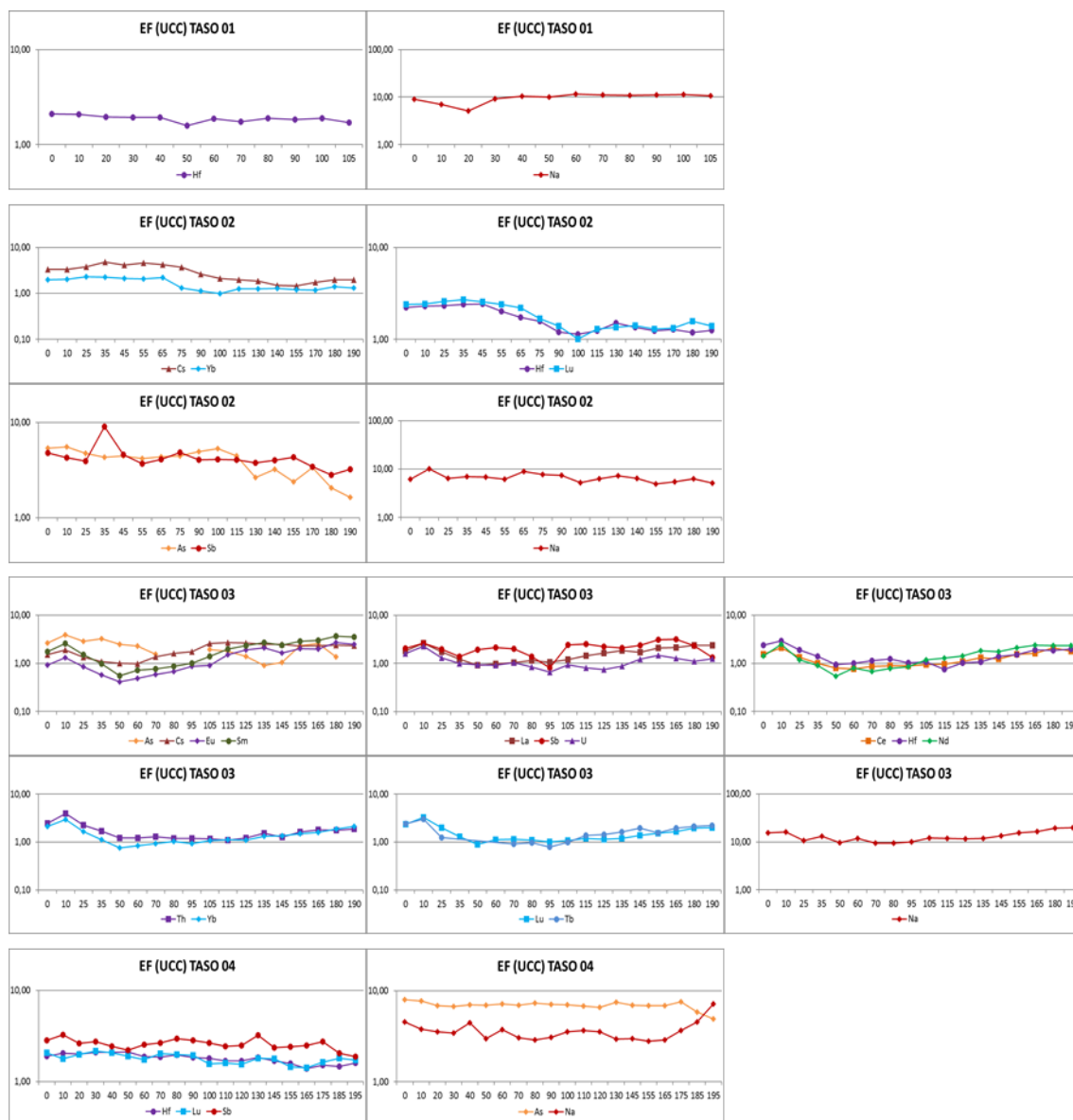


Figure 7: EF values for chemical elements in TASSO 01, TASSO 2, TASSO 3 and TASSO 4 profiles.

3.2 Sediment

3.2.1 Gamma spectrometry

Fig.8 shows the natural radionuclides ^{226}Ra , ^{210}Pb , ^{228}Ra , ^{228}Th and ^{40}K activity concentrations obtained by gamma spectrometry, as well as the radionuclides ^{238}U and ^{232}Th activities concentrations obtained by INAA for the sediment cores TASE 01, TASE 02, TASE 03 and TASE 04.

It was observed that, in all sediment cores, the highest activity concentrations were obtained for ^{210}Pb , with the highest values in TASE 04 and the lowest concentrations were obtained for ^{226}Ra in TASE 01, TASE 02 and TASE 03, and ^{228}Ra in TASE 04. The natural radionuclide ^{40}K was not determined.

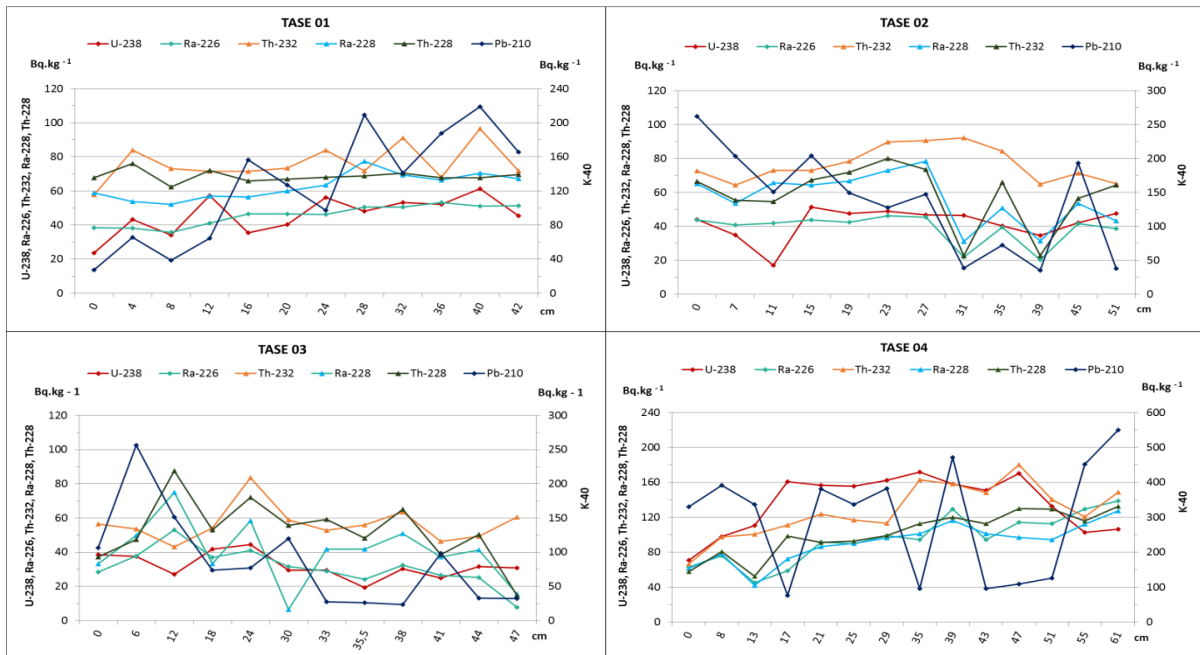


Figure 8: Concentrations of the natural radionuclides ^{238}U , ^{226}Ra , ^{210}Pb , ^{232}Th , ^{228}Ra and ^{228}Th , obtained in TASE 01, TASE 02, TASE 03 and TASE 04 sediment cores.

3.2.2 Instrumental neutrons activation analysis (INAA)

The chemical elements concentrations in the sediment cores TASE 01, TASE 02, TASE 03 and TASE 04 are shown in Figs. 9, 10, 11, and 12, respectively.

TASE 01, Fig. 9, showed a slight increase in the concentration of the elements Sb, Co, Hf, Sc, Rb, Zn and K up to 10 cm depth, and then a constant concentration as observed for the fine fraction of the sediment. For the element Fe the concentration decreased with depth and the elements Eu, Tb, Sm, Th, Nd, Ce and La showed a significant increase in their concentrations at depth 12 cm. TASE 02, Fig. 10, showed an increase in the concentrations of the elements Se and Cs and a reduction in the concentrations of the elements Eu, Sm, Nd, Zn and Fe with depth. The elements Sb, U, Yb, Se, Sm, La, As, Na and K showed a decrease of their concentrations at depth 11 cm.

TASE 03, Fig. 11, showed increase of the elements Ba, Na, K and As concentrations and reduction of the elements Ta, Tb, Sb, Eu and Zn concentrations with depth. A very high concentration for the element Zn when compared to the values of CCS (71 mg.kg^{-1}) [16] was determined mainly in the depth 12 cm, indicating a possible anthropic contribution due to the influence of the industries of the region in one of the tributaries rivers [19-20], located near the point of collection. TASE 04, Fig. 12, showed increase in the concentrations of the elements Lu, Co, Cs, Ta, Yb, Se, Rb, Th and Cr with depth, reduction in the concentration of elements As and Na with depth, and almost constant concentrations for the other elements.

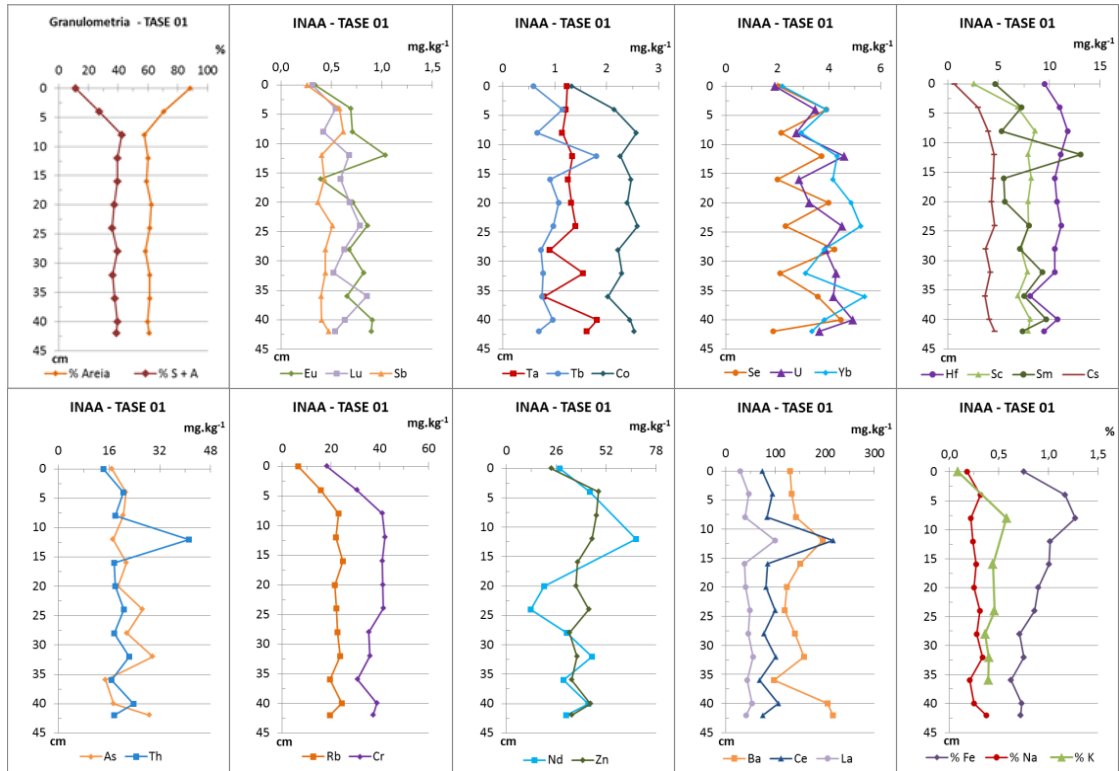


Figure 9: Concentrations of the elements obtained by INAA in TASE 01 sediment core.

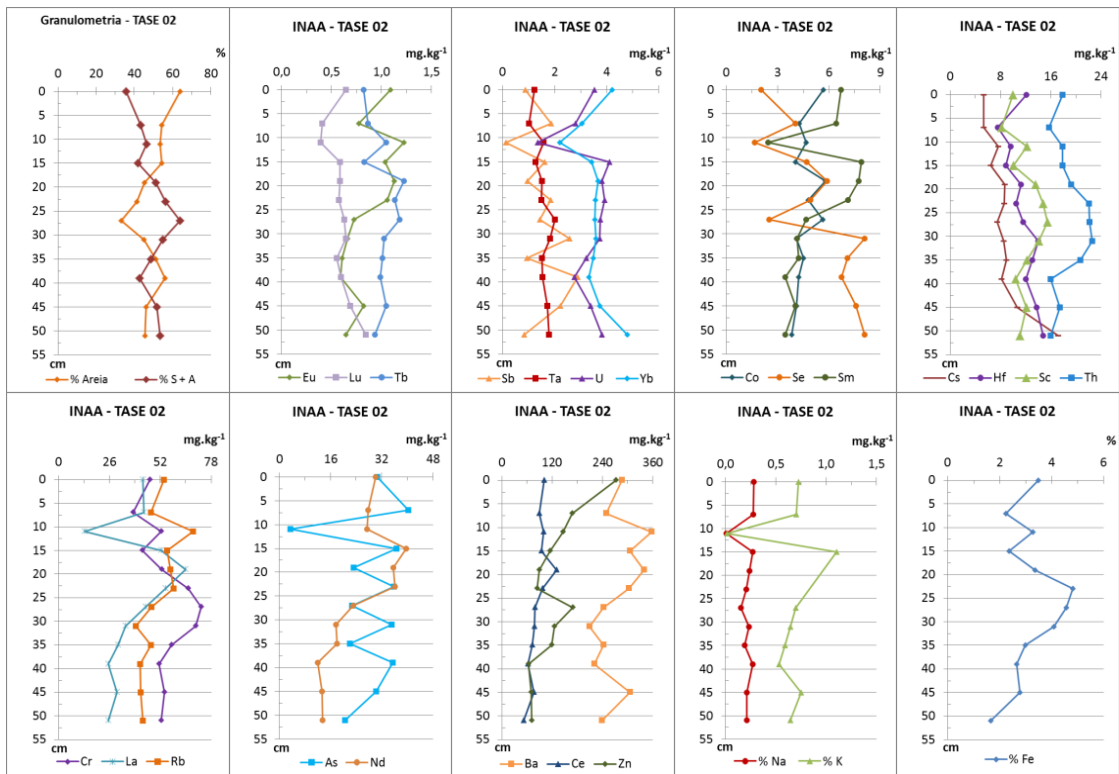


Figure 10: Concentrations of the elements obtained by INAA in TASE 02 sediment core.

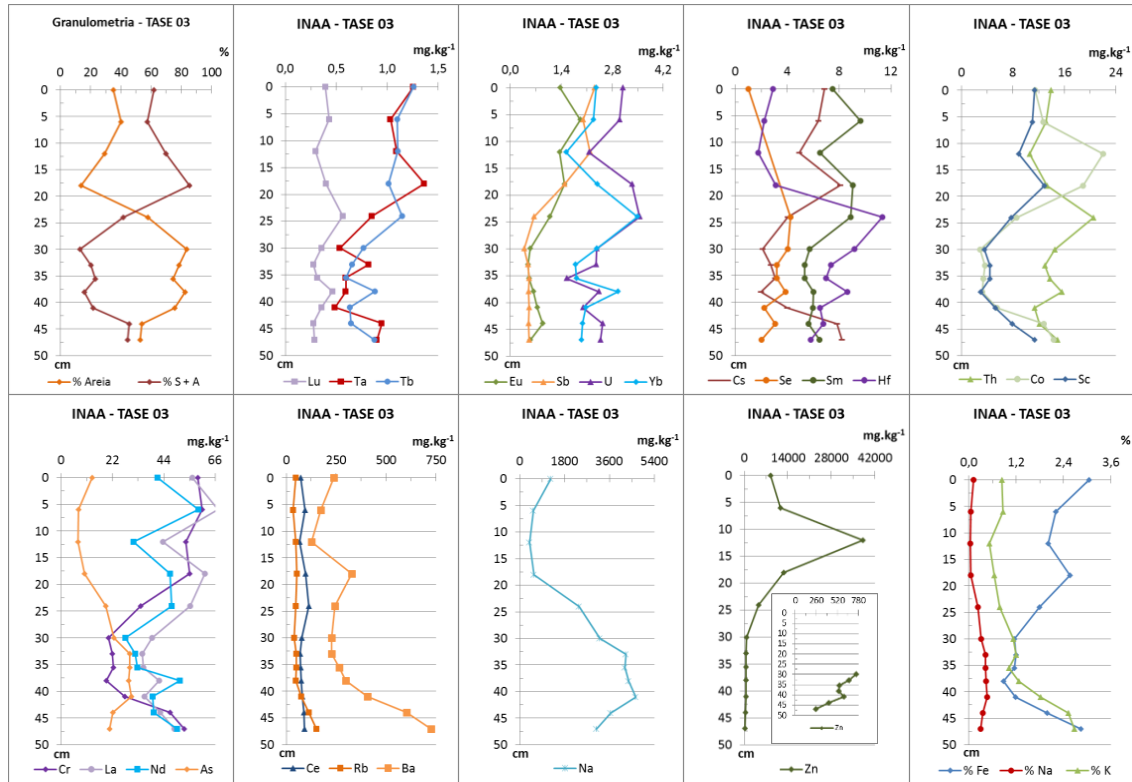


Figure 11: Concentrations of the elements obtained by INAA in TASE 03 sediment core.

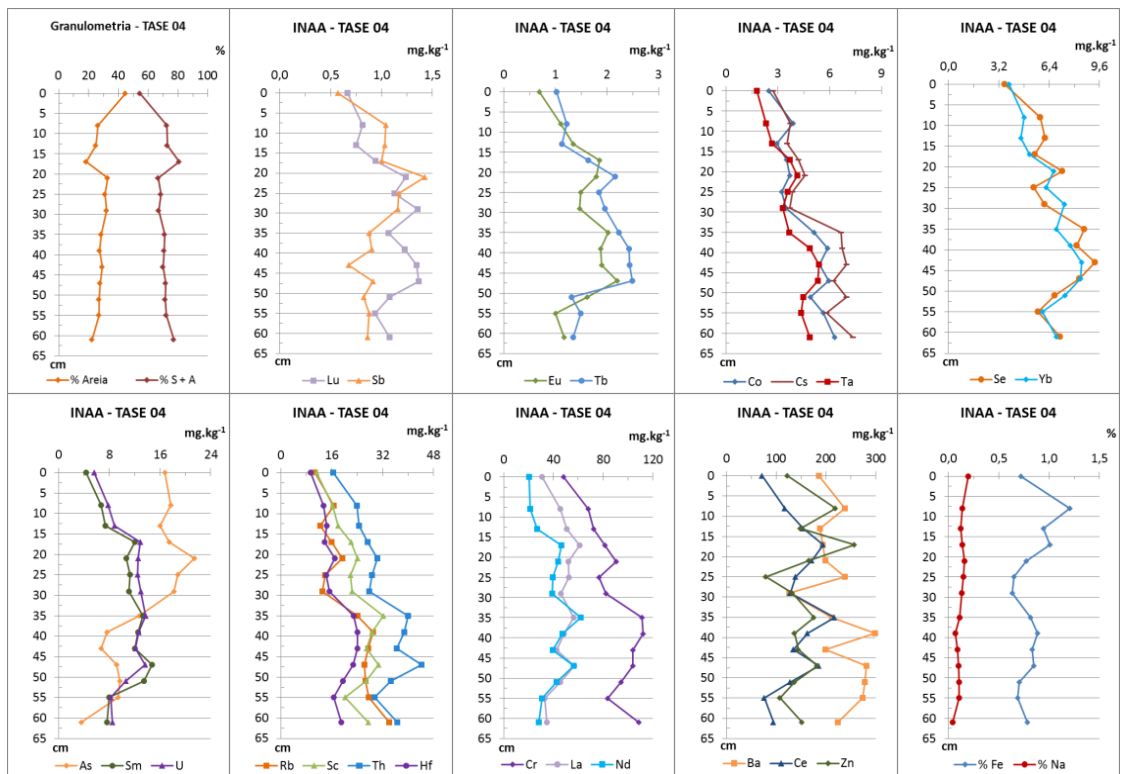


Figure 12: Concentrations of the elements obtained by INAA in TASE 04 sediment core.

3.2.3 Enrichment factor – EF

The results obtained of EF for TASE 01, TASE 02, TASE 03 and TASE 04 sediment cores are shown in Figs. 13 and 14.

TASE 01 presented very severe enrichment for the elements Na and As, very high enrichment for the elements Ce, Hf, Sb and Th and moderate for the other elements. TASE 02 showed very severe enrichment for the element Na at the beginning of the core and very high enrichment at the end; very high enrichment for the elements As and Sb, and moderate enrichment for the other elements.

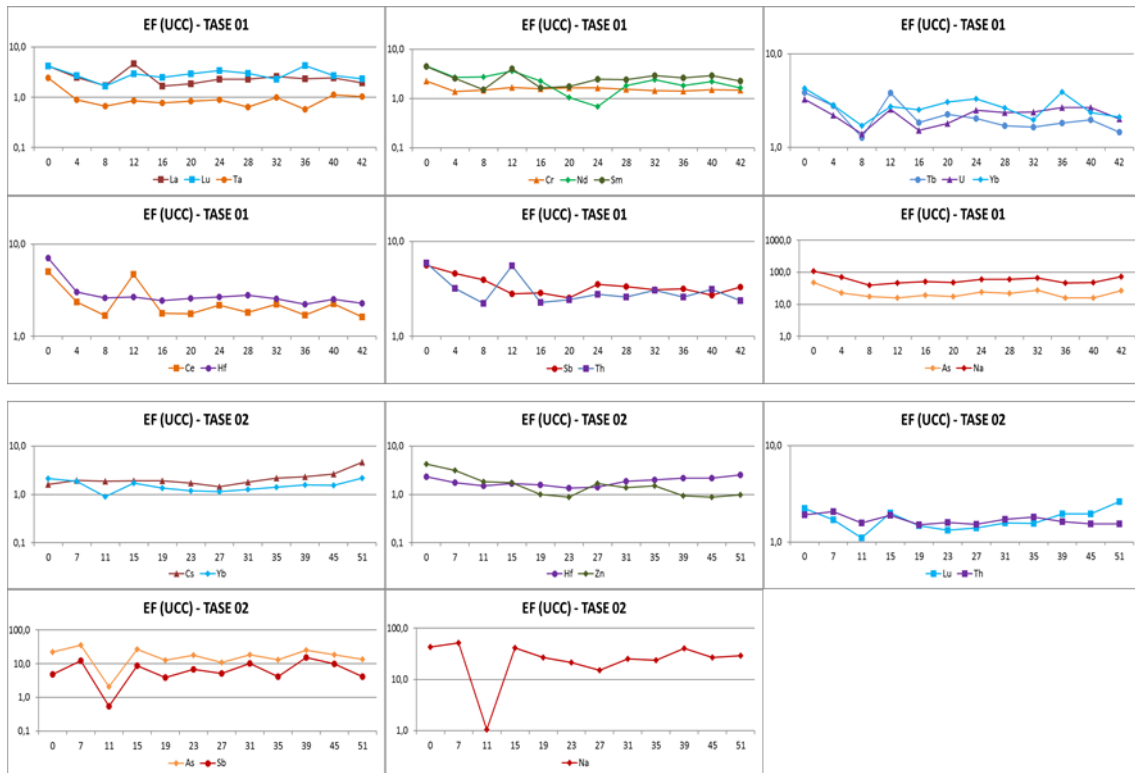


Figure 13: EF values for chemical elements in TASE 01 and TASE 02 sediment cores.

TASE 03 showed very severe enrichment for the elements Na, As at the middle and end of the core and Zn at the top; very high enrichment for Sb at the top of the core and for the elements Hf, La, Lu, Nd, Th and Yb at depth 38 cm. TASE 04 core showed very high enrichment for the elements Na and As at the beginning of the core and moderate at the end; moderate enrichment for the element Sb and U, throughout the core.

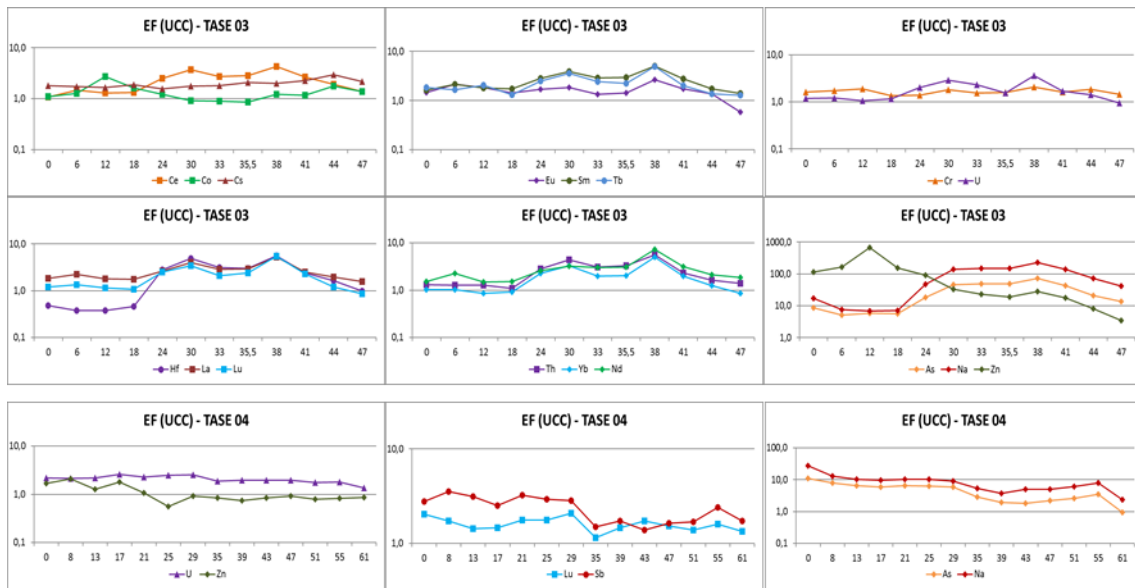


Figure 14: EF values for chemical elements in TASE 03 and TASE 04 sediment cores.

4. CONCLUSION

In this work four soil profiles and four sediment cores collected in the influence area of Taiacupeba reservoir were studied for the concentrations of ^{238}U , ^{226}Ra and ^{210}Pb radionuclides from the ^{238}U decay series, ^{232}Th , ^{228}Ra and ^{228}Th radionuclides from the ^{232}Th decay series, ^{40}K and the inorganic chemical characterization; the enrichment factor was also evaluated.

In the soil profiles the highest concentrations were obtained for the radionuclide ^{40}K in the majority of the samples and for ^{228}Th and ^{232}Th in TASE 03 profile and the lowest for ^{210}Pb . For the sediment cores it was verified that the highest concentrations were obtained for the radionuclide ^{210}Pb , mainly in the sediment core TASE 04 and the lowest for ^{226}Ra in the sediment cores TASE 01, TASE 02 and TASE 03, and ^{228}Ra in TASE 04.

On the inorganic chemical characterization of the soil and sediment of the reservoir, it was concluded that it presents high values for the elements Na, As and Sb, implying in continuous, regular and spatial monitoring. The sediment core TASE 03 presented a very high concentration of the element Zn when compared to the values of the Upper Continental Crust, indicating a possible anthropic contribution from the region's industries.

In the evaluation of the enrichment factor it was concluded that the soil presented very high enrichment for the elements Na (all profiles), As (TASE 02 and TASE 04) and Sb (TASE 2). The sediment presented very severe enrichment for the elements Na (TASE 01 and TASE 02), As (TASE 01 and TASE 03), and Zn (TASE 03); very high enrichment for the elements Ce, Sb and Th (TASE 01), As and Sb (TASE 02), Sb, Hf, La, Lu, Nd, Th and Yb (TASE 03), Na and As (TASE 04); and moderate enrichment for the other elements.

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