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Determination of Soil Screening Levels for natural radionuclides in Minas Gerais State, Brazil

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ABSTRACT

Soil screening levels express the levels of contaminant concentration in the soil, which guide the actions to be taken following investigation to confirm contamination. The list of toxic substances or elements under Brazilian legislation includes organics, volatile organics and metals but does not consider radioactive elements. Radioactive elements are all potentially carcinogenic and therefore need to be subject to legal control. The National Nuclear Energy Commission, the federal agency currently responsible for legislation regarding the control of Naturally-Occurring Radioactive Material (NORM) facilities does not establish guiding values for intervention in terms of soil activity concentration in the case of contamination with radioactive elements. In mining, the processing and treating of ores such as cassiterite, uranium, phosphate, niobium, and rare earths contribute to the generation of large amounts of NORM residues. Obviously, the improper disposal of these materials may lead to situations that result in soil and groundwater contamination and unnecessary exposure of the population in general. In order to establish guiding values for soil quality for natural radionuclides in the state of Minas Gerais, the study area included the entire state, which has unique characteristics related to the lithology, genesis, and morphology of the soils. These characteristics have tremendous influence on the petrogeochemistry of elements and radionuclides. A total of 110 soil samples were collected and analyzed in order to determine the activity concentration of U, Th, ^{226}Ra, ^{228}Ra e ^{210}Pb. In general, it was possible to verify that the activity concentrations of U are higher than those of Th. This fact can be explained by the intense weathering that most of the state's soil has undergone and the chemical and geochemical characteristics of the two elements. The values obtained up to the present are higher than the reference values for soil quality adopted in other parts of Brazil and the world.

1. INTRODUCTION

The Earth's crust contains radionuclides that are a major source of Naturally-Occurring Radioactive Materials (NORM) in the environment. The majority of these radionuclides belong to the decay chains that begin with ^{235}U, ^{238}U, and ^{232}Th.

When the radioactive nuclides associated with natural material come about after industrial processes, the material is called TENORM, Technologically Enhanced Naturally-Occurring

Radioactive Material. Mining and processing of ore for metal production generates large amounts of solid waste, around 1.5 billion tons per year, 100 million tons of which are metallurgical slag (Pontedeiro, 2006). The immense volume of NORM waste generated each year has drawn the attention of domestic and international environmental agencies as well as other regulatory agencies.

Inadequate disposal of this NORM/TENORM waste in the soil can cause problems such as damage to human health, degradation of the quality of water resources, restrictions to soil usage, damage to public and private property, and reduction of property values, as well as environmental damage. The contamination of the soil with radionuclides of natural series has become an even more serious problem since they have a long half-life and most are carcinogenic elements (Peres, 2007).

Management of contaminated areas seeks to minimize risks to which the population and environment are exposed by means of a set of measurements that ensure the areas and their impacts are known. They provide the instruments needed to make decisions about the most appropriate form of intervention.

In order to facilitate the decision making process for management of contaminated areas, establish priorities and reduce costs, environmental agencies from Brazil and several other countries have opted to establish soil value guidelines to be used in the first step of the decision making process. These are in turn defined as concentrations of a specific substance in the soil or groundwater that identify a soil as clean (CETESB, 2001).

Minas Gerais is an important historical reference in Brazilian cassiterite production, also standing out as a precursor of the metallurgical production of tin in the town of São João Del Rey in the 1940s. Cassiterite deposits have a genetic affinity with the Eastern Pegmatite Province of Minas Gerais, associated with columbite-tantalite and djalmite. It has been the target of intensive prospecting since the beginning of the 1940s.

For this reason, studies that contribute to establishing criteria and value guidelines for prevention and control of potentially toxic chemicals in the soil and groundwater are extremely important to the management of contaminated areas in the state of Minas Gerais.

2. PROCEDURES

2.1. Definition of the study area

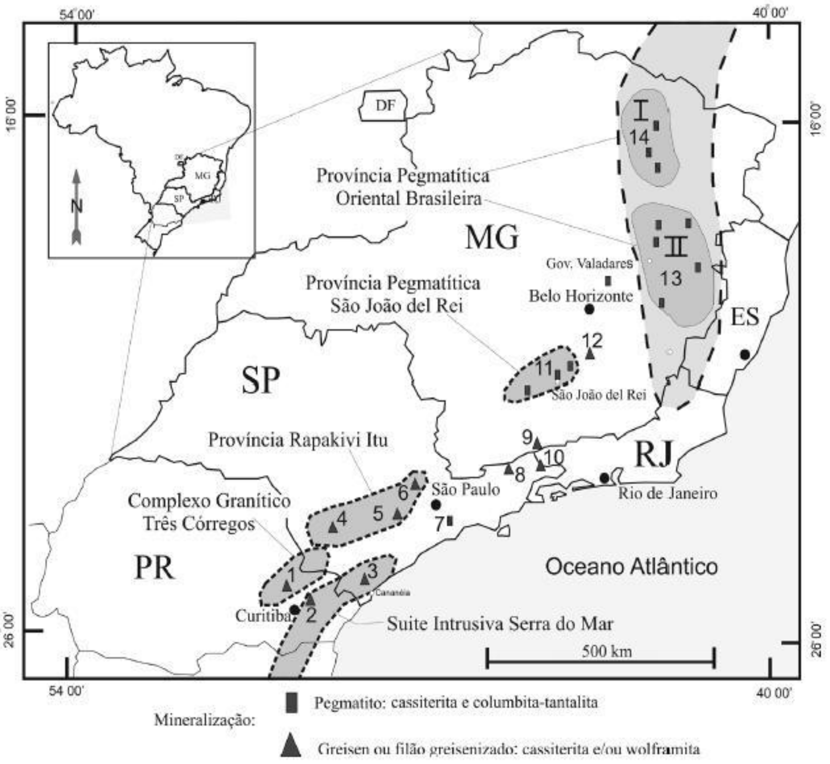
This study encompasses the whole state of Minas Gerais, which is located in south-eastern Brazil. It is the country's fourth-largest state, with an area of 588,384 km^2, equivalent to 7% of the nation's territory. The state has the third-largest economy in the country.

The history and traditions of the state of Minas Gerais are closely tied to mining activities and its enormous mineral reserves. The state extracts more than 160 million tons of iron ore, 29% of the country's mineral production, 53% of metallic mineral production, and around 50% of the country's gold production.

Uranium and thorium can occur in many types of rock. Three have the largest distribution: 1) bands of pre-Cambrian rocks; 2) areas with permeable sediment; and 3) areas with detrital sand accumulation.

In Minas Gerais, deposits can be considered large that have peculiar characteristics related to lithology, genesis, and morphology of soil that has significant influence on the pedogeochemistry of trace elements, metals, and radionuclides.

Figures 1 and 2 show the locations of large ore occurrences in the state of Minas Gerais.



Eastern Pegmatite Province; São João Del Rei Pegmatite Province; Rapakivi Itu Province; Três Córregos Granite Complex; Serra do Mar Intrusive Suite; Atlantic Ocean

Deposit: Pegmatite: cassiterite and columbite-tantalite; Greisen of greisenized vein: cassiterite and/or wolframite

Figure 1. Map of some pegmatite occurrences in the state of Minas Gerais (adapted from PPeGeo, the electronic Geosciences periodical portal).

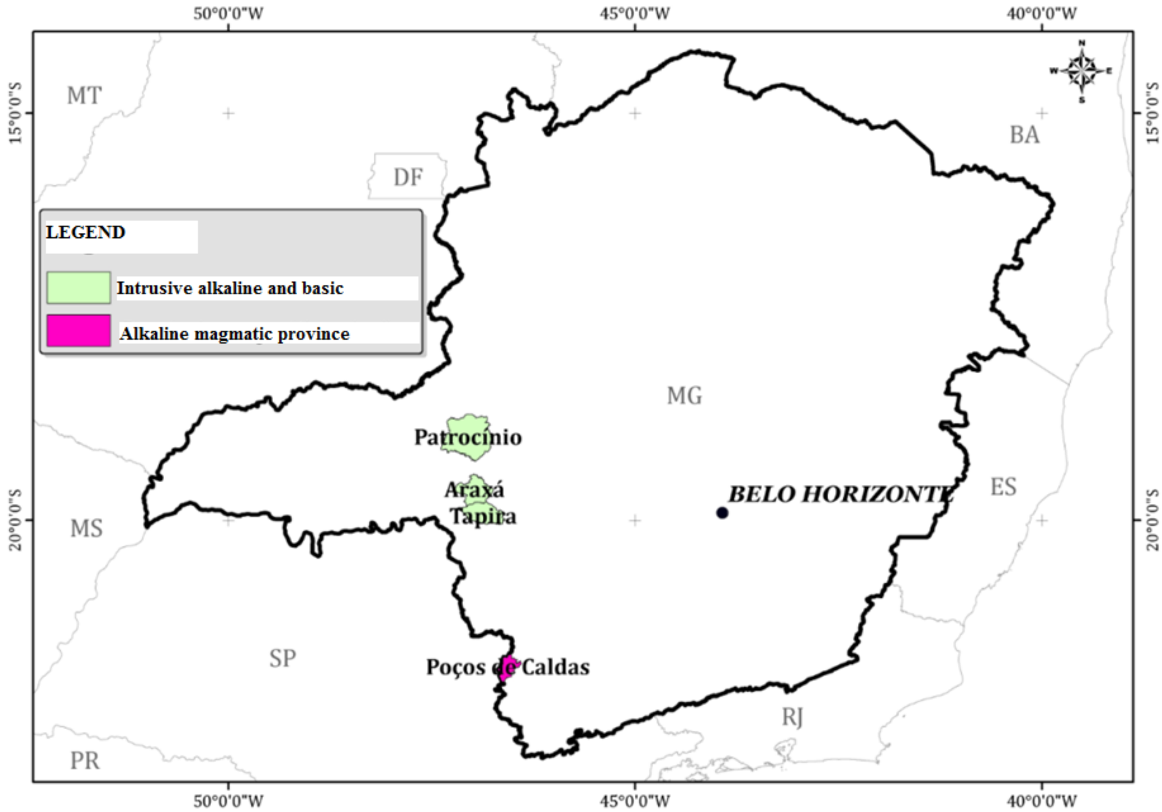


Figure 2. Map of some occurrences of intrusive alkaline and basic rocks in the state of Minas Gerais.

2.1.1. Sampling and analytical method

Samples were randomly collected within the map unit, observing the predomination of preserved or minimally impacted vegetation such as natural pastures. Other criteria were utilized to confirm the mapping unit, such as observation of soil profiles in highway excavations, erosions, and gullies, as well as interpretation of vegetation and geomorphology. The soil samples were taken from a depth of 0 to 20 cm, which is equivalent to horizon A for most soil.

For natural radionuclides analysis, 110 samples were chosen from the 500 that are in the Soil Database by superimposing existing information about soil classes and the geology of each region and trying to obtain greater reach and make them more representative within the analytical limitations. 12 of the samples taken were tripled in order to make sure that the U, Th, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{40}K analysis and preparation method could be reproduced.

The points chosen for U, Th, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{40}K analysis are well distributed throughout Minas Gerais as can be seen in Figure 3.

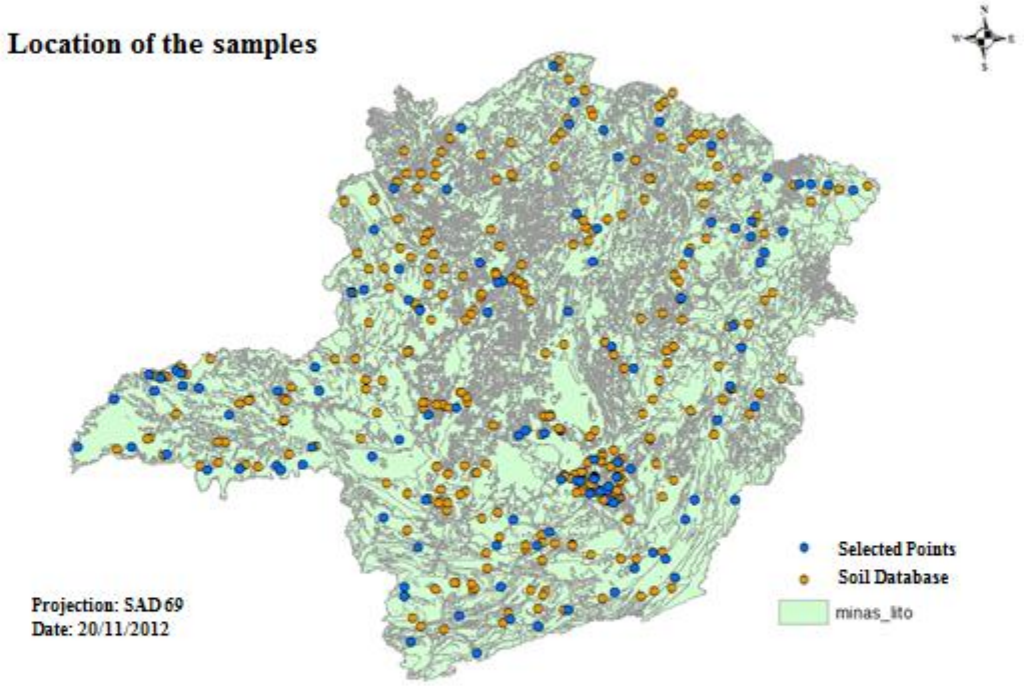


Figure 3. Location of the samples chosen from the state of Minas Gerais Soil Database to determine soil quality reference values for U_{nat} and Th.

The UV-VIS molecular absorption spectrometry method (Savvin, 1961) was used to determine uranium and thorium concentration in the soil samples. This method was chosen because it is simple, accessible and inexpensive when compared to other uranium-determination methods. Pre-concentration and extraction steps were employed to increase the detection limit and analytical sensitivity.

The method was validated by means of IAEA reference 327, *Radionuclides in Soil*. Table 1 shows the methods' Minimum Detection Limits for U_{nat} and Th.

The Gamma spectrometry was used to to determine the ^{40}K, ^{226}Ra, ^{228}Ra and ^{210}Pb concentration in the soil samples. This method was chosen because it is a rapid method for obtaining information several radionuclides, including ^{40}K, ^{226}Ra, ^{228}Ra and ^{210}Pb. The advantages of using gamma spectrometry with germanium detector are due primarily to its resolution time (about 10-8 s), their linearity response over energy wide range, the activities of a large number of radionuclides can be determined simultaneously, besides being a non-destructive technique. It is a good method precision for analysis of natural radionuclides in environmental samples (Siqueira, 2009).

Table 1. Minimum Detection Limits of the Methods.

Radionuclide	LD ($BqKg^{-1}$)
U	0.51
Th	0.27
^{226}Ra	20
^{228}Ra	20
^{210}Pb	30
^{40}K	90

3. RESULTS AND DISCUSSION

3.1 U and Th concentration results

Figure 4 shows the results for U concentration distributed by yield class on the petrologic map; Figure 5 shows the results for Th.

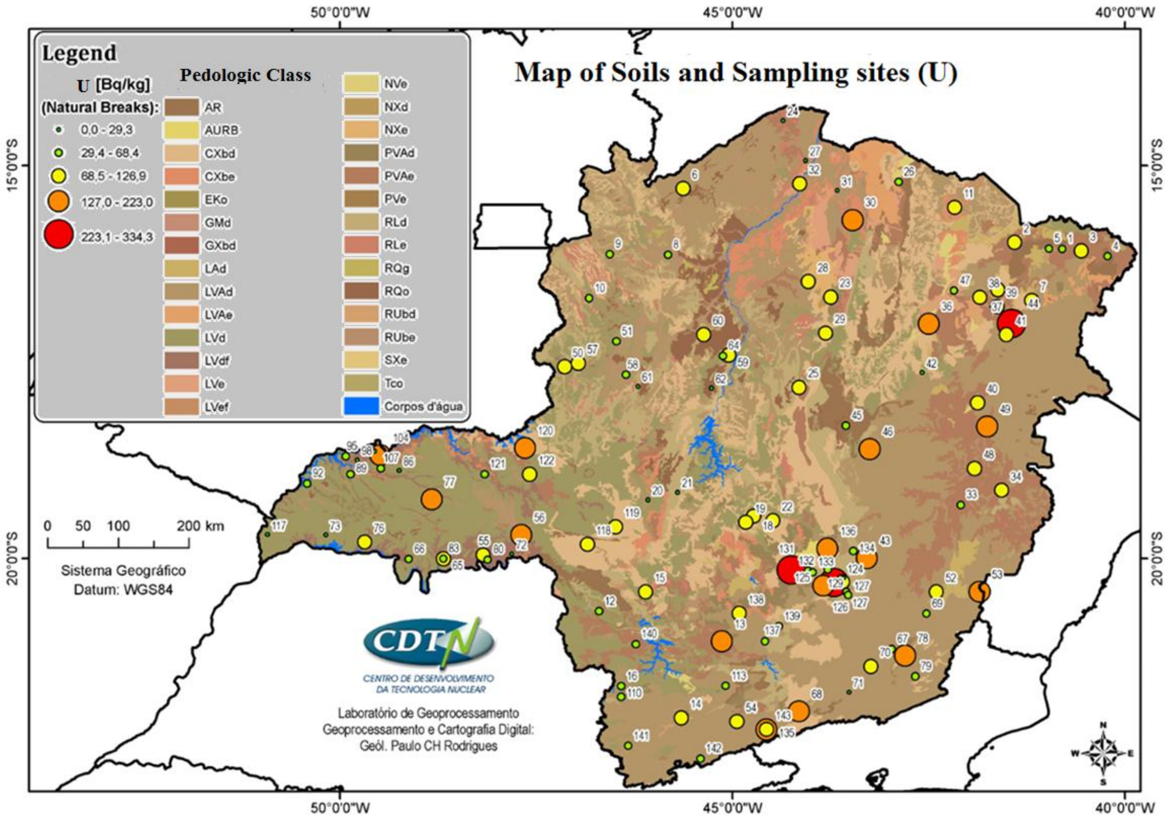
The Natural Breaks method was used to cluster numerical data into U and Th concentration yield classes (Jenks, 1967). This method identifies the limits of each class. The Natural Breaks method is a data classification method that determines the best arrangements of values in different classes and seeks to reduce variance within classes and maximize variance between classes.

The concentration level variation obtained can be visualized from its spatial distribution on the map.

As can be seen in Figures 4 and 5, there is a large variation in U and Th concentrations. From spatial observation, there seems to be no clear correlation between concentration levels for the two elements.

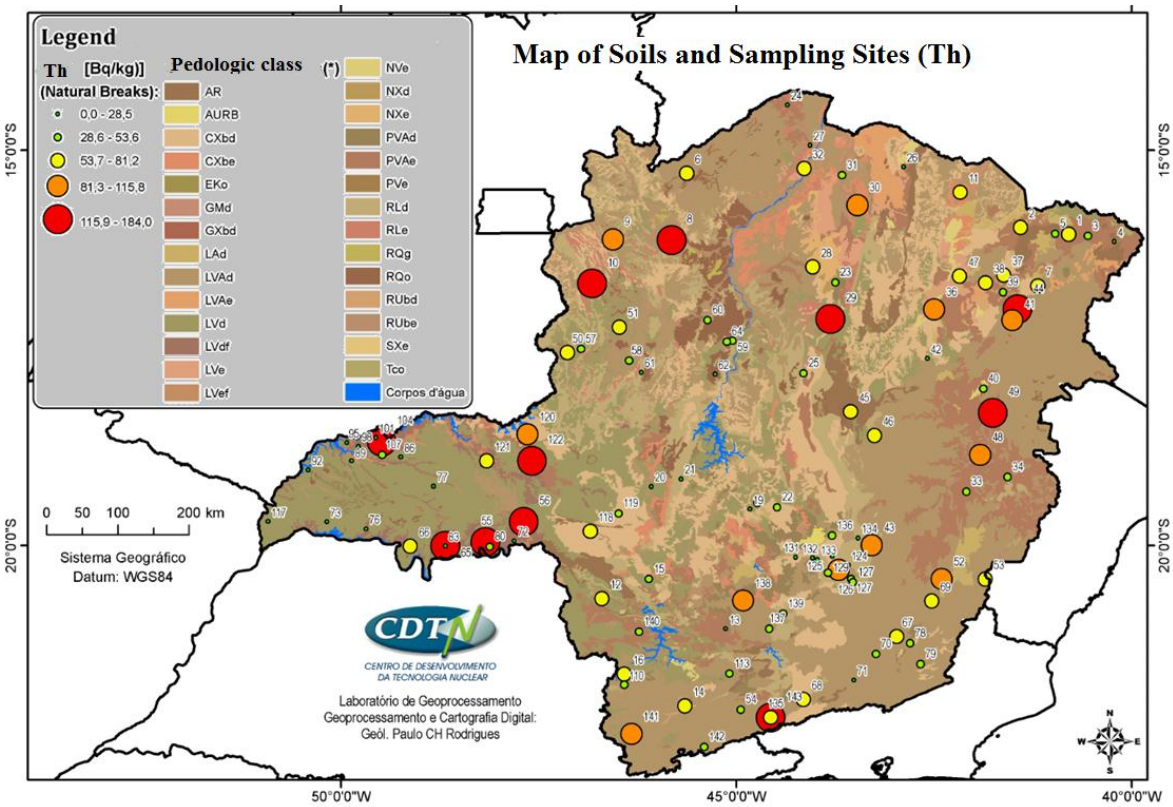
High U and Th concentration values were found in regions where the elements are common, for example: the pegmatite provinces in the northeast of the state; the alkaline magmatic provinces of Poços de Caldas; the basic and alkaline intrusive in Tapira, Araxá and Salitre; the migmatite bands in the São João Del Rei and Itabirito regions; as well as the detrital sand accumulation at Rio das Mortes in São João Del Rei.

In order to isolate the most relevant and stable structures and standards identified by the set of object data, a descriptive analysis of the U, Th, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{40}K concentration data set was carried out. The results are shown in Figures 6 to 11.



Soil and sample point map (U); Legend; Uranium; Petrologic class; Bodies of water; Geographical system; Geoprocessing laboratory; Geoprocessing and digital cartography; Decimal points in table, eg, 0.0 – 29.3

Figure 4. U_{nat} concentration distributed by U_{nat} concentration level classes ($Bq.kg^{-1}$).



Soil and sample point map (Th); Thorium; Decimal points

Figure 5. Th concentration distributed by Th concentration level classes (Bq.kg^{-1}).

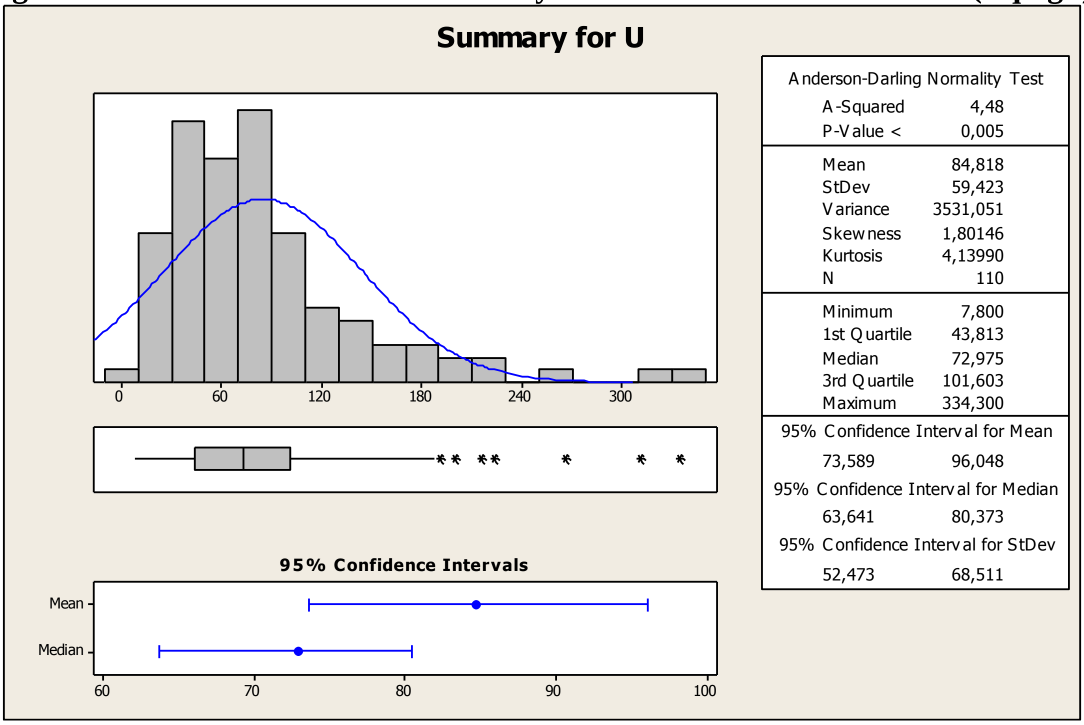


Figure 6. Descriptive analysis of U results

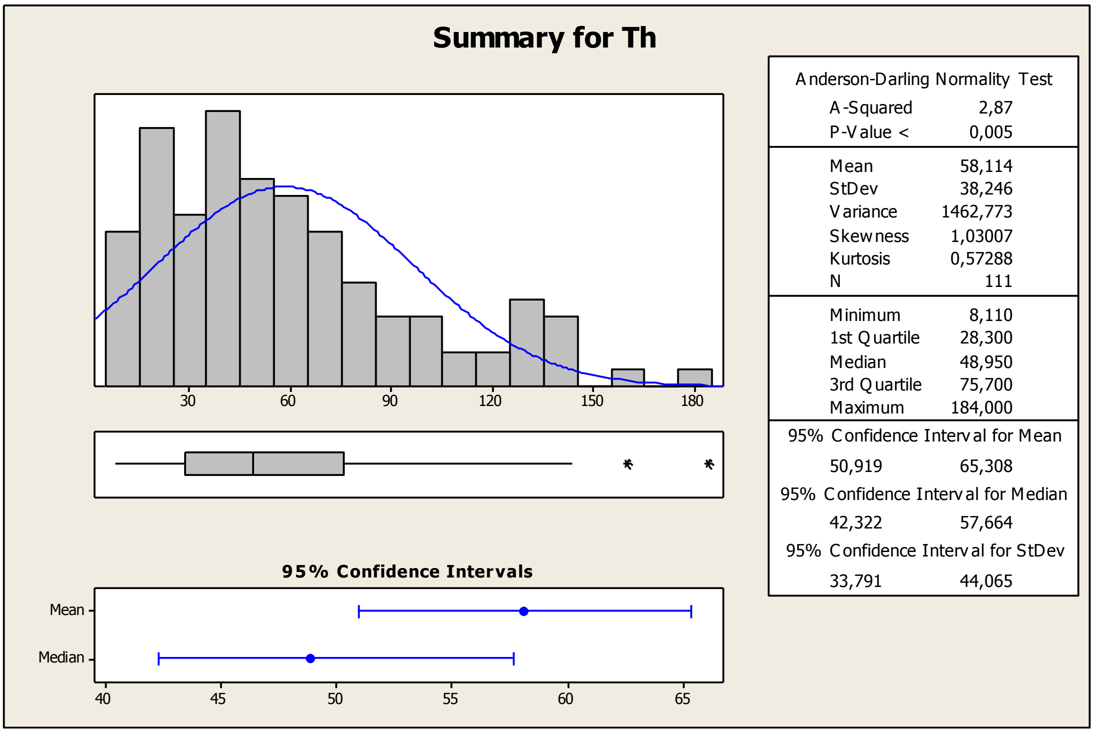


Figure 7. Descriptive analysis of Th results.

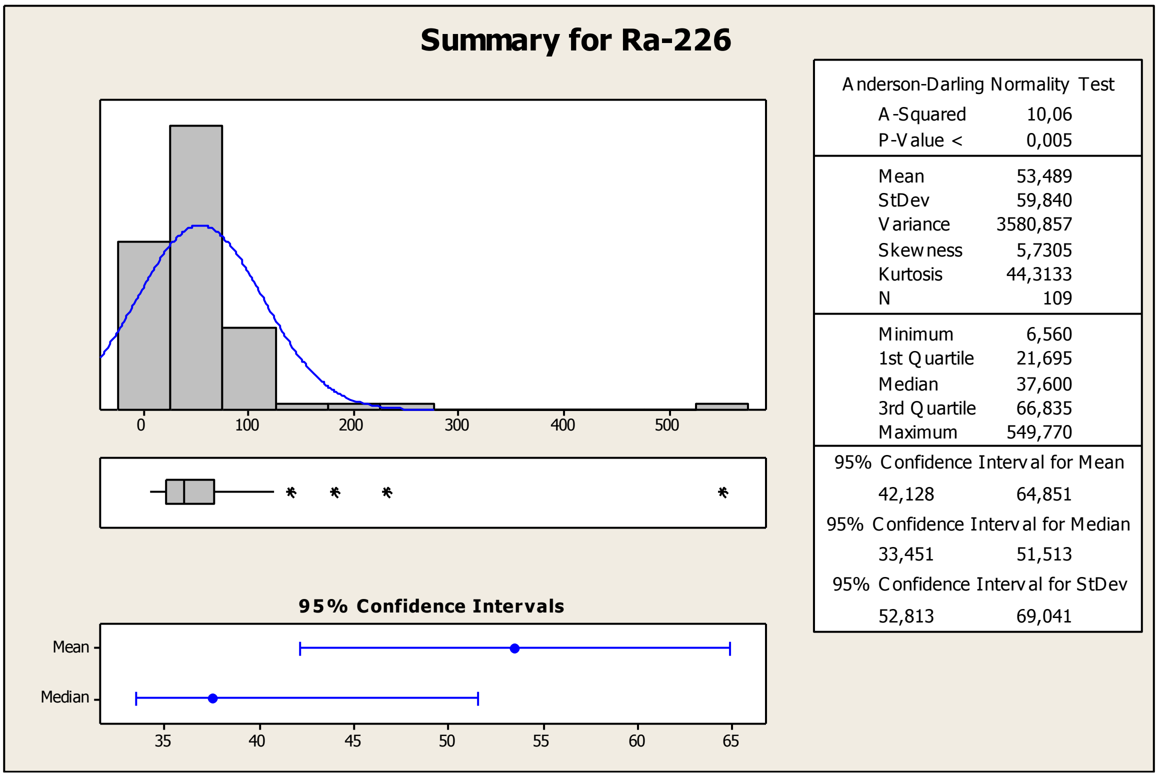


Figure 8. Descriptive analysis of ^{226}Ra results.

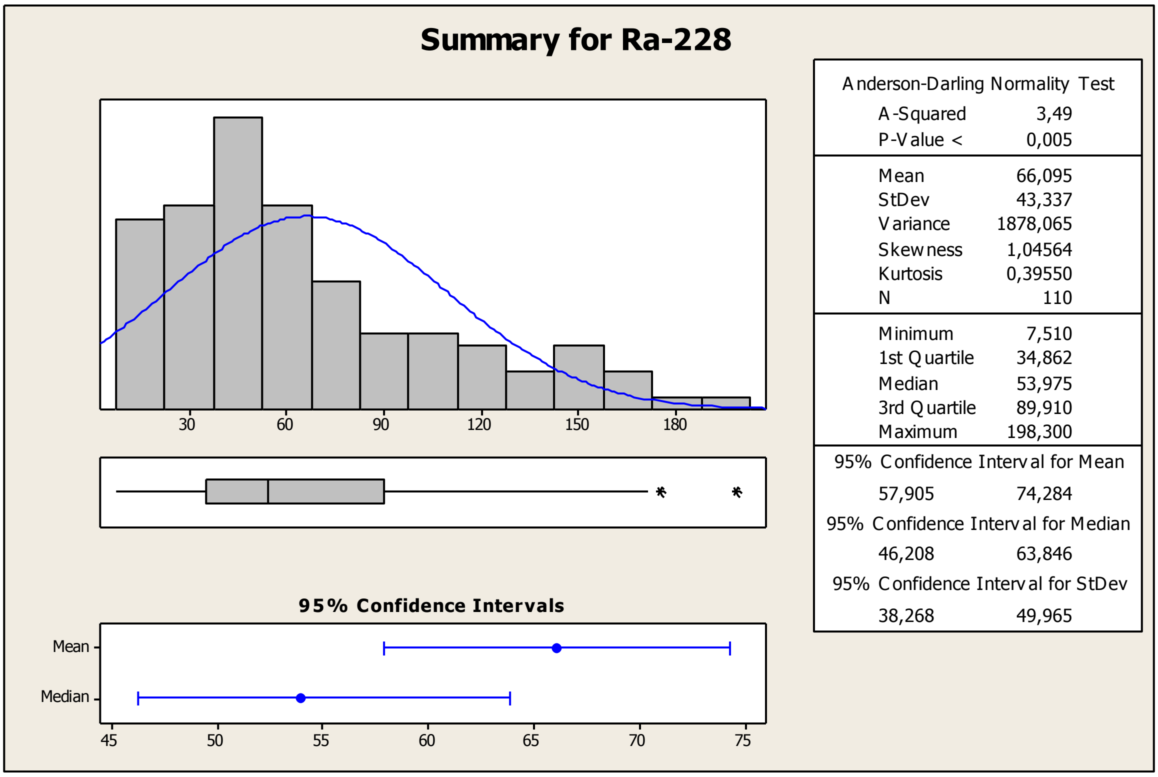


Figure 9. Descriptive analysis of ^{228}Ra results.

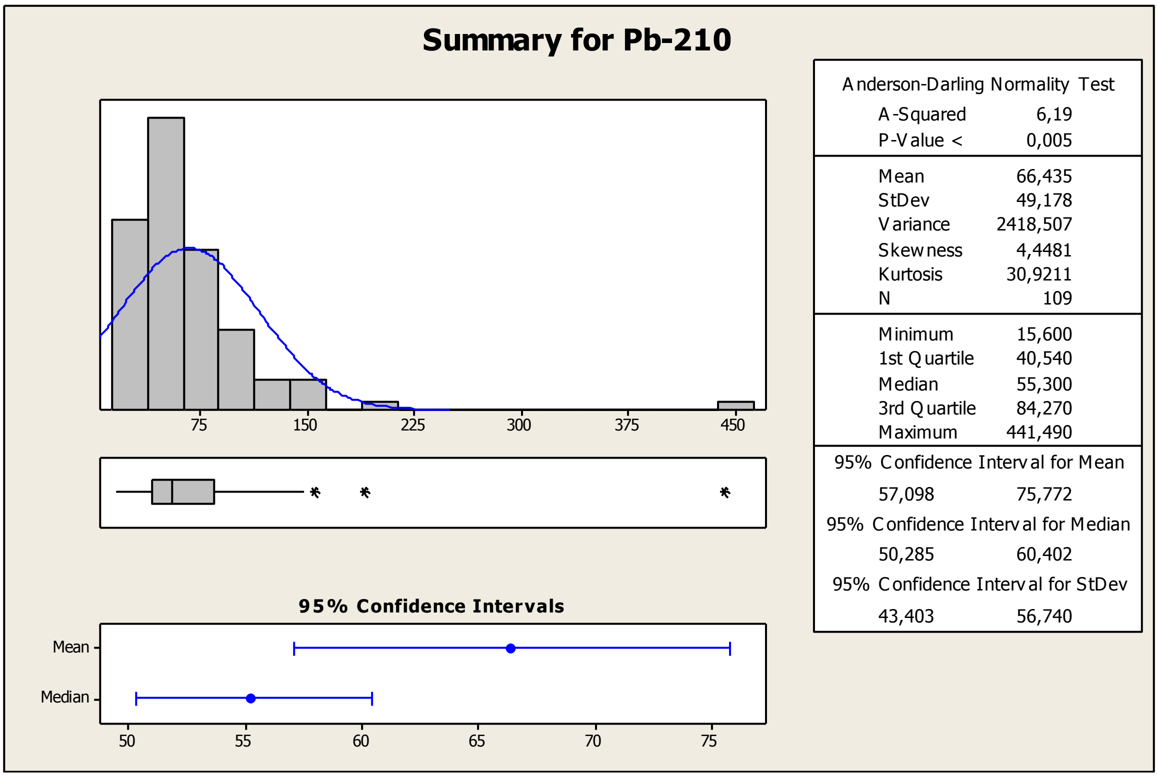


Figure 10. Descriptive analysis of ^{210}Pb results.

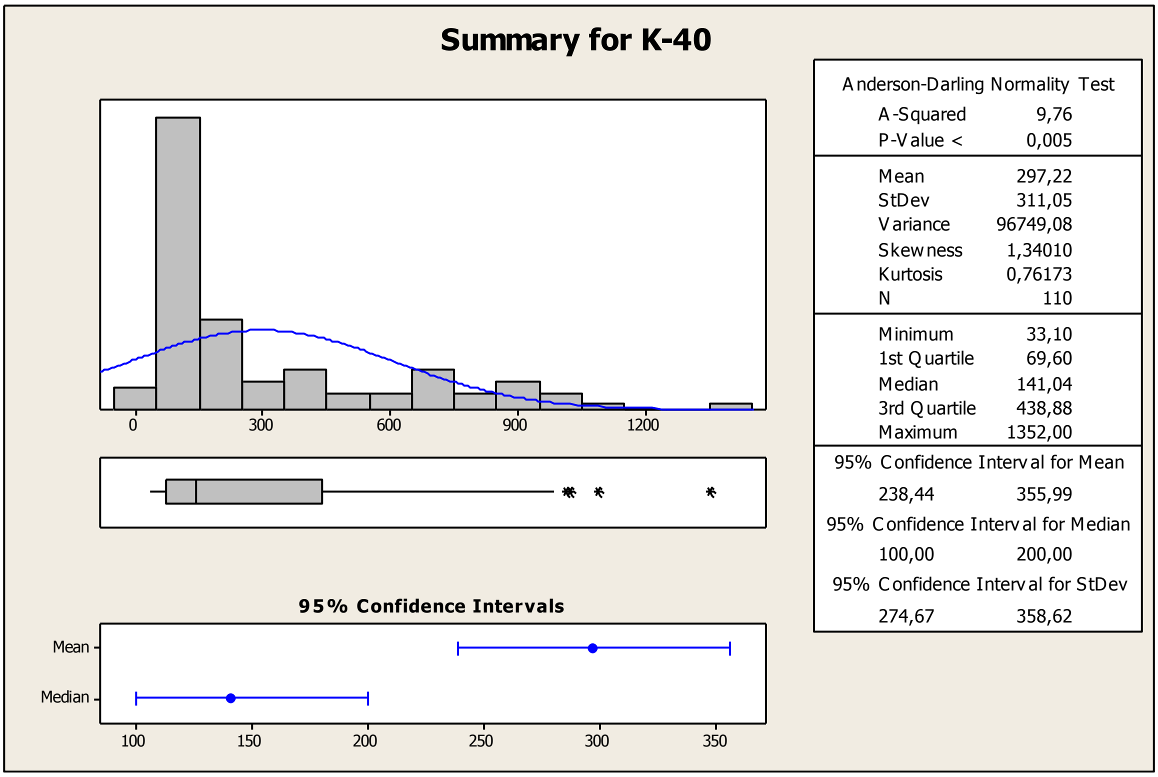


Figure 11. Descriptive analysis of ^{40}K results.

3.2 - Soil quality reference values

Soil quality reference values (QRV) for natural radionuclides in soils in the state of Minas Gerais were based on statistical processing of the data applied in the two sets of concentration results.

Table 2. Quality Reference Values (QRV) for natural radionuclides in soils in the state of Minas Gerais.

Radionuclide	Quality Reference Value ($Bq.kg^{-1}$)
U	101.6
Th	75.7
^{226}Ra	66.8
^{228}Ra	89.9
^{210}Pb	84.3
^{40}K	438.9

3 - CONCLUSIONS

QRVs were determined based on statistical processing of concentration results. Proposed values were 101.6 $Bq.kg^{-1}$ for U, 75.7 $Bq.kg^{-1}$ for Th, 66.8 $Bq.kg^{-1}$ for ^{226}Ra, 89.9 $Bq.kg^{-1}$ for ^{228}Ra, 84,3 $Bq.kg^{-1}$ for ^{210}Pb and 438.9 $Bq.kg^{-1}$ for ^{40}K. These values are higher than both those found in the state of São Paulo (Peres, 2007) and world average concentration values of Th (30 $Bq.kg^{-1}$), ^{238}U (35 $Bq.kg^{-1}$) and ^{226}Ra (35 $Bq.kg^{-1}$),found in soil (UNSCEAR, 2000), indicating the importance of the geological characteristics of each region.

Note that the highest concentration values for both U and Th were seen in regions of Minas Gerais in which there are radioactive anomalies, such as: the pegmatite provinces in the northeast of the state; the alkaline magmatic provinces of Poços de Caldas; the basic and alkaline intrusive in Tapira, Araxá and Salitre; the migmatite bands in the São João Del Rei and Itabirito regions; as well as the detrital sand accumulation at Rio das Mortes in São João Del Rei.

Generally speaking, U concentrations in the soil were higher than Th concentrations. This can be explained by the weathering process that most of the soils in the state of Minas Gerais have undergone as well as the chemical and geochemical characteristics of the two elements.

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