Joel Augusto Moura Porto, Lander de Jesus Alves, Hector Hugo Silva Medrado, Josilene da Silva Rocha, Rodrigo Gibaut de Souza Góis, Fábio Carvalho Nunes, Marcelo Machado Viana, Ana Paula de Carvalho Teixeira

https://doi.org/10.1590/SciELOPreprints.13524

Submetido em: 2025-09-29
Postado em: 2025-11-12 (versão 1)
(AAAA-MM-DD)

Determination of Natural Uranium and Thorium Levels in Environmental Matrices Surrounding Uranium Mining Sites via ICP-MS

Joel Augusto Moura Porto

Department of Chemistry, Federal University of Minas Gerais (UFMG), Belo Horizonte, Minas Gerais, Brazil. ORCID: https://orcid.org/0000-0002-0121-1403

Lander de Jesus Alves

State University of Santa Cruz (UESC), Postgraduate Program in Biology and Biotechnology of Microorganisms, Ilhéus, Bahia, Brazil. ORCID: https://orcid.org/0000-0003-4448-6594

Hector Hugo Silva Medrado

Brazilian Nuclear Industries (INB) - Uranium Concentration Unit, Security, Radioprotection and Environment Management, Caetité, Bahia, Brazil. ORCID: https://orcid.org/0000-0003-4567-6699

Josilene da Silva Rocha

Brazilian Nuclear Industries (INB) - Uranium Concentration Unit, Security, Radioprotection and Environment Management, Caetité, Bahia, Brazil. ORCID: https://orcid.org/0009-0008-2196-2088

Rodrigo Gibaut de Souza Góis

Brazilian Nuclear Industries (INB) - Uranium Concentration Unit, Security, Radioprotection and Environment Management, Caetité, Bahia, Brazil. ORCID: https://orcid.org/0000-0003-2839-7532

Fábio Carvalho Nunes

Federal Institute of Education, Science and Technology of Bahia (IFBaiano), Santa Inês, Bahia, Brazil. ORCID: https://orcid.org/0000-0002-5954-397X

Marcelo Machado Viana

Department of Chemistry, Federal University of Minas Gerais (UFMG), Belo Horizonte, Minas Gerais, Brazil. ORCID: https://orcid.org/0000-0002-6682-2504

Ana Paula de Carvalho Teixeira*

Department of Chemistry, Federal University of Minas Gerais (UFMG), Belo Horizonte, Minas Gerais, Brazil. ORCID: https://orcid.org/0000-0002-4985-7502

*Correspondent author: anapaula.cta1@gmail.com

ABCTRACT

The link between energy use and environmental health is crucial, yet the role of thorium near uranium deposits remains poorly understood despite known issues with nuclear power. In this context, this study determines the concentration of natural thorium in environmental matrices around a uranium mining site andexplores the distribution of natural uranium and thorium radionuclides in ore from the Cachoeira and Engenho mines in Brazil, as well as in aqueous and soil samples, using ICP-MS. Despite distinct ore fingerprints, the Engenho mine exhibits a lower uranium-to-thorium ratio than the Cachoeira mine. However, thorium levels from both mines are significantly lower than those reported in the literature, where concentrations exceed 150 Bq.kg^{-1}. Conversely, thorium levels in aqueous samples from both mines are significantly higher than literature values, suggesting that uranium concentrations in these sites could be influenced by hydrological and geological conditions. The range of thorium levels in Cachoeira mine is 1.27 x 10^{4} to 1.8 x 10^{3} Bq.L^{-1}, but in Engenho mine, the values are 2.41 x 10^{4} to 3.69 x 10^{3} Bq.L^{-1}. The agricultural crop soil values, such as bean, corn, cassava, and prickly pear, are lower than the literature estimates, suggesting little to no input from nearby mines. The range of thorium activity in agricultural soil was 90.97 to 99.42 Bq.kg^{-1}, which is higher than the values reported in literature in areas without uranium mining (28 ± 39 Bq.kg^{-1}). Understanding the geographic variability of these radionuclides informs health policies and supports sustainable mining practices, promoting environmental and community safety.

Keywords:

Uranium, thorium, mining, ICP-MS, ore, water, soil.

1. Introduction

The interconnection between the natural environment and energy management represents an extremely relevant concern in the context of preserving the health of our planet [1,2]. The methods used in the acquisition and consumption of energy have the potential to generate considerable impacts on the environment, ranging from the emission of gases responsible for the greenhouse effect to the depletion of ecosystems [3,4]. Within this scenario, the need to seek energy sources that are characterized by their cleanliness and renewability emerges, with a view to ensuring a future marked by sustainability [5,6].

Nuclear energy is a form of energy obtained from reactions that release a significant amount of energy, making nuclear energy an attractive source for generating electricity and other applications [7]. One of the main characteristics of nuclear energy is its energy density, that is, the amount of energy released per unit of mass is much greater compared to other energy sources [8]. Furthermore, nuclear energy is considered a clean energy source in terms of greenhouse gas emissions, as it does not emit CO_2 during electricity generation [9,10].

The use of nuclear energy is controversial due to the risks associated with nuclear safety and the appropriate management of radioactive waste [11,12]. On the other hand, fossil fuels, such as oil, coal and natural gas, are non-renewable energy sources that have been widely used over the years [13]. However, burning it releases large amounts of greenhouse gases, contributing to climate change and other negative environmental impacts [14].

As its main raw material, uranium is mostly used in the production of nuclear energy [15]. Its most common isotope is U-235, which is fissionable and capable of sustaining a chain reaction [16]. Uranium extracted from the Earth's crust (isotopically rich in U-238) goes through an enrichment process, in which the concentration of U-235 is increased to allow nuclear reactions to be viable [17].

Despite presenting several advantages, such as high energy density, low greenhouse gas emissions and the availability of uranium as a raw material, there are some counterpoints to be considered, such as the risks associated with nuclear safety, adequate waste management of radioactive substances and the proliferation of nuclear weapons [18]. In this context, uranium mining is an activity intrinsically linked to the production of nuclear energy and, as such, faces specific issues related to health and safety in the workplace, as well as the socioeconomic implications in the surrounding areas [19].

It is natural that exposure to radioactivity is a constant concern for uranium mine workers, as they are at risk of inhaling radioactive particles, which increases, for example, the risk of lung disease and lung cancer [20,21]. To mitigate such risks, protective measures, such as the use of personal protective equipment, regular monitoring of radiation exposure and nuclear safety training, are fundamentally necessary [22].

The safety of communities near mines is also a significant concern, as uranium extraction can, to some extent, contaminate drinking water and soil, posing local health risks [23]. However, it is important to emphasize that uranium and thorium are naturally present in the Earth's crust, and their occurrence in rocks, soils, and groundwater in the region predates mining activities. Therefore, mining does not introduce these elements into the environment, but it can influence their mobility or concentration in specific environmental compartments.

To address these environmental, public health and industrial concerns, it is essential to know natural uranium (U-nat) and thorium (Th-nat) concentrations in ores and environmental samples close to uranium mines. The literature, however, points out the difficulties in determining radionuclides in samples from environmental matrices, especially species that are not as naturally abundant as others, such as aluminium, iron, calcium or sodium [24–26]. This implies, in addition to the difficulty of quantifying radioisotopes, the small range of matrices explored for this purpose [27,28]. Hyphenated analytical techniques, such as Inductively Coupled Plasma Mass Spectrometry (ICP-MS), have proven effective in accurately quantifying uranium concentrations, allowing for a more complete and detailed assessment of the risks and benefits associated with uranium mining [29–31].

In this context, the motivation for this study lies in the need to characterize the natural levels of uranium and thorium in the vicinity of a uranium mining company. The research aims to determine the concentrations of these elements in environmental matrices, including water, soil, and ore samples from the region. As the company operates two distinct mining sites, a comparative analysis will also be conducted to investigate potential variations in uranium and thorium content between them.

2. Methodology

To effectively establish the concentrations of uranium and thorium in the environment surrounding the uranium mining company, a systematic sampling method was employed. The sampling locations were selected based on their proximity to the two uranium mines: Cachoeira Mine (deactivated) and Engenho Mine (active). This will give a representative analysis of both mine environments and their potential impacts on surrounding ecosystems.

In addition, a comparison between the two mines will be done to contrast uranium and thorium concentration differences. The selected sampling points, descriptions, and Universal Transverse Mercator (UTM) coordinates are given in Tables 1 and showed in Figures S1, S2, and S3. Except for the ore samples, which were collected as early as 2012, all other samples were collected between February 2021 and December 2023.

It is also important to highlight that the ICP-MS analysis quantified specifically the isotopes Th-232 and U-238. For comparison with literature values commonly reported as “Th-nat” and “U-nat,” the measured concentrations were converted according to their natural isotopic composition. In the case of thorium, natural thorium is essentially composed of Th-232, so the concentration of Th-nat was taken as equal to the measured concentration of Th-232, i.e., $C_{Th\text{-}nat} \approx C_{Th\text{-}232}$. For uranium, natural uranium consists predominantly of U-238 (99.2745%), with minor fractions of U-235 (0.720%) and U-234 (0.0055%); thus, U-nat was obtained from the measured U-238 concentration according to $C_{U\text{-}nat} = C_{U\text{-}238}/0.992745$. This approximation is valid for natural, non-enriched materials, while in cases of isotopic fractionation or enrichment/depletion each isotope must be quantified separately.

2.1. SAMPLES COLLECTION

2.1.1. Ores

The collection of ore samples (before and after the leaching process) was carried out in conjunction with the mining unit's ore quality control team. Ore samples were collected from assembled leach piles. Subsequently the samples were crushed to particle sizes in the range of 2 to 1.2 mm.

2.1.2. Surface and Subterranean Water

The aqueous samples were collected from designated points at the Cachoeira and Engenho mines. Depending on the water source, samples were obtained from pits or wells using a submersible pump or a bailer for wells with low water volume. Approximately 1 liter of water was collected per sample.

2.1.3. Soils

For the collection of soil samples, the cleaning conditions of the hoe was initially checked and it was cleaned using water. Then, a superficial cleaning was carried out in the area intended for collection, in order to remove any foreign objects, such as stones, sticks, roots and leaves.

Then, an area of approximately 0.2 m² was demarcated using a wooden template. The collection itself was carried out carefully, reaching a maximum depth of 10 centimeters. After collection, the sample was homogenized and divided into four equal parts. Approximately 2 kg of each sample was placed in a plastic bag, ensuring that it contains information regarding the date and point of collection. To preserve the integrity of the sample, it was necessary to close the plastic bag immediately after obtaining the sample, using a string.

In addition to points directly associated with mining areas, point LR036A was included, located downstream of the planned Engenho mine pit area, with the purpose of characterizing surface drainage in a future mining zone.

For comparative purposes, soil samples were also collected from agricultural areas at least 20 km away from the mines (beans, corn, cassava, and prickly pear crops). These soils were considered reference samples, representing the natural background of the region in areas used for agriculture and livestock.

2.2. SAMPLES PREPARATION

2.2.1. Filtration of Aqueous Samples

The filtration procedure for aqueous samples was carried out with a vacuum system with a vacuum flask using sterilized cellulose nitrate filter membranes with a porosity of 0.45 µm. This procedure is essential for some samples that are very muddy. The filtered volume for each sample intended for analysis of natural uranium was 50 mL.

2.2.2. Drying of Solid Samples

Solid samples were dried in an oven at 130–200 °C for 12 hours. This temperature range was adopted in accordance with laboratory internal protocols to ensure complete removal of moisture and to avoid re-adsorption of uranium and thorium on wet particles, which could compromise subsequent acid digestion and ICP-MS quantification. We acknowledge that in environmental studies drying at room temperature is common; however, for this specific study, high-temperature drying was essential to guarantee reproducibility and minimize variations related to residual humidity.

Initially, the sample (~2.0 kg) was weighed using a precision portable balance. Then, the sample was carefully placed in an aluminium dish and then taken to the oven, where it was subjected to a temperature that varies between 130 ºC and 200 ºC, for a period of 12 hours. Every 3 hours, the tray was removed from the stove.

Table 1. Description and location in UTM coordinates of the collection points.

POINT	LATITUDE	LONGITUDE	DESCRIPTION
E004A	13°49'47.34"S	42°17'14.23"W	Collection point located in the pre-basin, which receives the liquids that flow through the interception channel at the base of the waste rock pile from the Cachoeira mine, from the rainwater that flows over the waste and leached ore deposit and that percolate through the tailing's piles of this deposit.
E004B	13°49'42.26"S	42°17'14.96"W	Collection point located in the fines sedimentation basin, which receives the waters that overflow from the pre-basin.
E006	13°50'54.54"S	42°16'48.83"W	Fines sedimentation basin of the Engenho mine.
E009	13°50'0.46"S	42°17'47.42"W	Collection point located inside the mine pit 1 of Cachoeira mine.
E012	13°50'2.58"S	42°17'56.74"W	Collection point located inside the mine pit 2 of Cachoeira mine.
ES08	13°49'50.91"S	42°17'22.68"W	Collection point located in the interception channel at the base of the waste rock pile, whose waters originate from pumping of water accumulated in the Cachoeira mine pits, flow over the piles of the Waste and Leached Ore Deposit and percolation in the piles of this deposit.
ES08M	13°49'53.41"S	42°17'27.44"W	Mining at the basis of the slope of the waste and leached ore deposit, whose contribution is only drainage waters that percolate the waste and leachate piles, without influence from the mine.

ES17D	13°51'17.86"S	42°16'38.96"W	In pit sump of mine pit II - reservoir built within an excavation or mining pit to collect and manage water, used to control and drain excess water from the mining operation to prevent flooding and maintain safety and operational efficiency.
ES18D	13°51'8.47"S	42°16'57.87"W	In pit sump of mine pit III - reservoir built within an excavation or mining pit to collect and manage water, used to control and drain excess water from the mining operation to prevent flooding and maintain safety and operational efficiency.
LR036A	13°50'50.19"S	42°16'32.15"W	Surface water/sediment sampling point located in a drainage 700 m downstream of the planned site of the Engenho mine pits.
FG08	13°51'19.65"S	42°16'35.06"W	Geotechnical drilling hole – Engenho mine.
FG09	13°51'22.75"S	42°16'33.55"W	Geotechnical drilling hole – Engenho mine.
S001			Composite sample from each pile of ore after the leaching process, from the sampling carried out in the pile.
S002			Composite sample of each pile of ore during assembly, before the leaching process, based on sampling carried out in the pile.

At this point, the sample was carefully mixed with a spatula, ensuring uniform drying. After the sample has completely dried, it was removed from the oven and placed on the bench to cool. A new weighing was carried out to obtain the dry weight of the sample. After waiting around 1 hour for the sample to reach approximately room temperature, it was transferred to a 60# mesh sieve, placed inside a mortar. At this point, the sample was subjected to a sieving process, with careful back and forth movements.

2.2.3. Sample Preparation by Microwave-Assisted Digestion

The digestion of solid samples for the analysis of natural uranium was carried out based on the methodologies proposed by Krug [32] and Vieira [33].

For the digestion process, a Microwave-Accelerated Reaction System (MARS) was used, model MARS 6®, applied in laboratories for digestion, dissolution, hydrolysis, extraction or drying of a large scale of materials [34,35]. Its primary purpose is for rapid sample preparation for atomic absorption and inductively coupled plasma, emission spectroscopy, and gas or liquid chromatography [36]. The digestion process used for each solid matrix is presented in the following topics.

2.2.4. Digestion of ore samples

The ore sample, weighing 0.5 g, was placed in Teflon- Perfluoroalkoxy alkane (PFA) digestion tubes for microwave digestion with 10 mL of concentrated HF and HNO_3. After that, a pre-digestion period of 20 minutes was waited. After pre-digestion was completed, the Teflon tubes were symmetrically distributed in the carousel according to the number of tubes. A single-step acid digestion program was employed. The temperature was ramped up to 180 °C over 15 minutes, followed by a 15-minute hold at that temperature.

After digestion in the digester, 0.5 mL of H_2O_2 was added to the tubes, waiting approximately 5 minutes. The resulting supernatant was transferred to a 250 mL Teflon beaker. In general, at the end of this stage, there was still solid material deposited at the bottom of the tube or material with a gelatinous consistency. In this case, 10 mL of 4% (w/v) H_3BO_3 were added to the tube containing the sample, followed by a new digestion in the microwave digester.

The supernatant resulting from the second digestion was transferred to the same Teflon beaker that contained the supernatant from the first digestion. This liquid was placed on a hot plate at 280°C until completely dry. Subsequently, 20 mL of concentrated HNO_3 and 1 mL of H_2O_2 were added to the dry residue, returning to the hot plate at 280°C until complete drying to ensure the total elimination of HF. After complete drying, 70 mL of 2% (v/v) suprapure HNO_3 solution was added with a 100 mL PP beaker. Finally, the mixture was again placed on the hotplate at 280°C until the volume reached 25 mL.

2.2.5. Digestion of soil samples

Soil analysis also requires the digestion of 0.5 g of sample. And unlike the mineral digestion process, soil digestion rarely presented a gelatinous appearance after leaving the sample from the microwave digester. In this sense, it was not necessary to use 10 mL of 4% (w/v) H_3BO_3 in the intermediate stage of the digestion process. The rest of the process is analogous to that presented in ore digestion.

A two-step acid digestion program was applied. In the first step, the temperature was ramped to 200 °C over 15 minutes and held for 15 minutes. In the second step, an additional 15-minute ramp was followed by a 15-minute hold, also at 200 °C.

2.3 Chemical Analyses

Given its great sensitivity and precision in measuring trace elements, ICP-MS is the method we chose for determining natural uranium and thorium. This technique has the advantage of being able to measure uranium and thorium in complicated matrices, such as mining samples, once the signal intensity is unaffected by the molecular environment of the element being measured [37].

ICP-MS has been effectively used to analyze uranium in different matrices, such as environmental samples, high-purity gadolinium sulfate, and human urine. [38–42]. Likewise, ICP-MS has demonstrated its effectiveness in determining thorium also in different matrices, such as titanium, urine, and environmental samples [43–45].

In this study, natural uranium and thorium were determined using an inductively coupled plasma mass spectrometer PerkinElmer NexION® 300X ICP-MS equipped with a collision cell. Measurements were performed using both helium (He) collision mode (White Martins-Praxair, Sertãozinho, SP, Brazil) and no gas mode. Table S1 shows the Operating parameters adopted for PerkinElmer NexION® 300X ICP-MS used in the analyses for uranium and thorium.

2.3.1 Calibration and Parameters

To construct the calibration curve for uranium and thorium analyses, standard solutions were used: for uranium, a 5 $\mu g \cdot L^{-1}$ solution prepared from a Specsol® standard; and for thorium, a standard solution from Agilent® with a concentration of 1001 $\mu g \cdot L^{-1}$. Both standards were prepared in 5% HNO_3. The different concentrations referring to the points of the calibration curve were generated with the sampler and automatic dilutor coupled to the ICP-MS equipment, C-2 DX Autosampler (Optima ELAN Version).

Calibration curves for uranium and thorium are shown in Figures S4 and S5, respectively. The reagents were of Chemically Pure (CP) quality and the water used to bulk or dilute was ultrapure. The working solutions, prepared to be used in the analysis of blanks, standards and samples, were obtained from reagents stored in a solubilized liquid state. The internal standard used was Elemental Scientific M1-ISMS-25 prepFAST ICPMS Internal Standard 8000 ppb (In-115).

The limits of detection (LOD) obtained for ICP-MS were 0.01 $\mu g \cdot L^{-1}$ (0.25 $Bq \cdot L^{-1}$) for U-238 and 0.005 $\mu g \cdot L^{-1}$ (0.02 $Bq \cdot L^{-1}$) for Th-232, ensuring sufficient sensitivity for quantification in the environmental samples analyzed.

2.3.2 Quality Assurance and Validation

The analytical methodology for thorium determination was specifically developed and optimized in our laboratory to meet the requirements of environmental monitoring associated with uranium mining. As part of the validation process, we highlight the use of the certified reference sample PNI 463 from the National Intercomparison Program for the Analysis of Environmental Samples for Radionuclide Determination (PNI – IRD/CNEN, Brazil), which was included in the analytical batches.

The same PNI protocol was also applied to uranium determinations, ensuring consistency and traceability of results. In addition, our laboratory participated in an intercomparison round (TO21/0423 for Th-232 and TO26/0423 for U-238) conducted while these environmental samples were being analyzed, further confirming the accuracy and reliability of the methodology (Supplementary Material 1).

3. RESULTS AND DISCUSSION

The analyses are presented here by different matrices. In each topic, a discussion will initially be made for each category and, at the end, a comparative analysis will be made between radionuclides and between the two mines: Cachoeira and Engenho.

3.1. Ore Samples

3.1.1. Natural Uranium

Some samples of crushed ores from the Cachoeira and Engenho mines were analyzed for their natural uranium content (Figure 1A). In Figure 1A, the points labeled S002 represent crushed ore samples, and S001 refers to the leachate. The letter C indicates samples from the Cachoeira mine, while E denotes the Engenho mine.

The leaching process begins with the crushed ore being stacked in piles. A concentrated solution of sulfuric acid is then applied to the ore. This acid solution facilitates the extraction of uranium from the ore by breaking down the minerals that contain uranium [46]. Over time, this process disrupts the secular equilibrium between the parent nuclides (Uranium-238, for example) and their decay products (the "children", such as Thorium-230, Radonium-222, and others) [47]. In an undisturbed uranium deposit, this equilibrium remains stable for long periods [48]. However, the leaching process disrupts this balance, as the acid helps release the uranium into the solution, which is then collected as leachate (S001). This process of leaching allows for the uranium to be separated from the rest of the ore, making it easier to recover and process.

One of the most striking observations is the much higher uranium concentration in the Cachoeira mine. Taking average values, the crushed ore from Cachoeira (S002C) has 49,530 Bq/kg, whereas Engenho's crushed ore (S002E) has 25,010 Bq/kg, nearly half as much, showing that uranium deposits in Cachoeira are naturally more concentrated. The same trend is observed in the leachate, in which Cachoeira (S001C) has 9,950 Bq/kg, while Engenho (S001E) has only 4,300 Bq/kg.

Another important point is the variability in the data. Cachoeira's uranium concentrations show greater fluctuations, as indicated by its higher standard deviations: 4,806 Bq/kg for crushed ore and 445.4 Bq/kg for leachate. This points to the fact that uranium is not evenly distributed throughout the ore, leading to a more heterogeneous deposit. Engenho's standard deviations, in contrast, are much lower (977.3 Bq/kg for crushed ore and 106.3 Bq/kg for leachate), which indicates a more uniform composition in this mine.

To assess the efficiency of uranium extraction, we examined the ratio between uranium concentration in leachate (S001) and crushed ore (S002). In the Cachoeira mine, this ratio is 0.2009, meaning that, on average, 20.09% of the uranium from the crushed ore is transferred to the leachate. In the Engenho mine, the ratio is 0.1720, indicating an extraction efficiency of about 17.20%.

The Student's t-test applied to the natural uranium concentrations in the two mines yielded a two-tailed p-value of 0.0066, which is well below the conventional significance threshold of 0.05. This indicates that the difference in uranium concentrations between the Cachoeira and Engenho mines is statistically very significant, making it highly unlikely that the observed variation is due to random sampling. In practical terms, this confirms that Cachoeira has a naturally higher uranium concentration than Engenho.

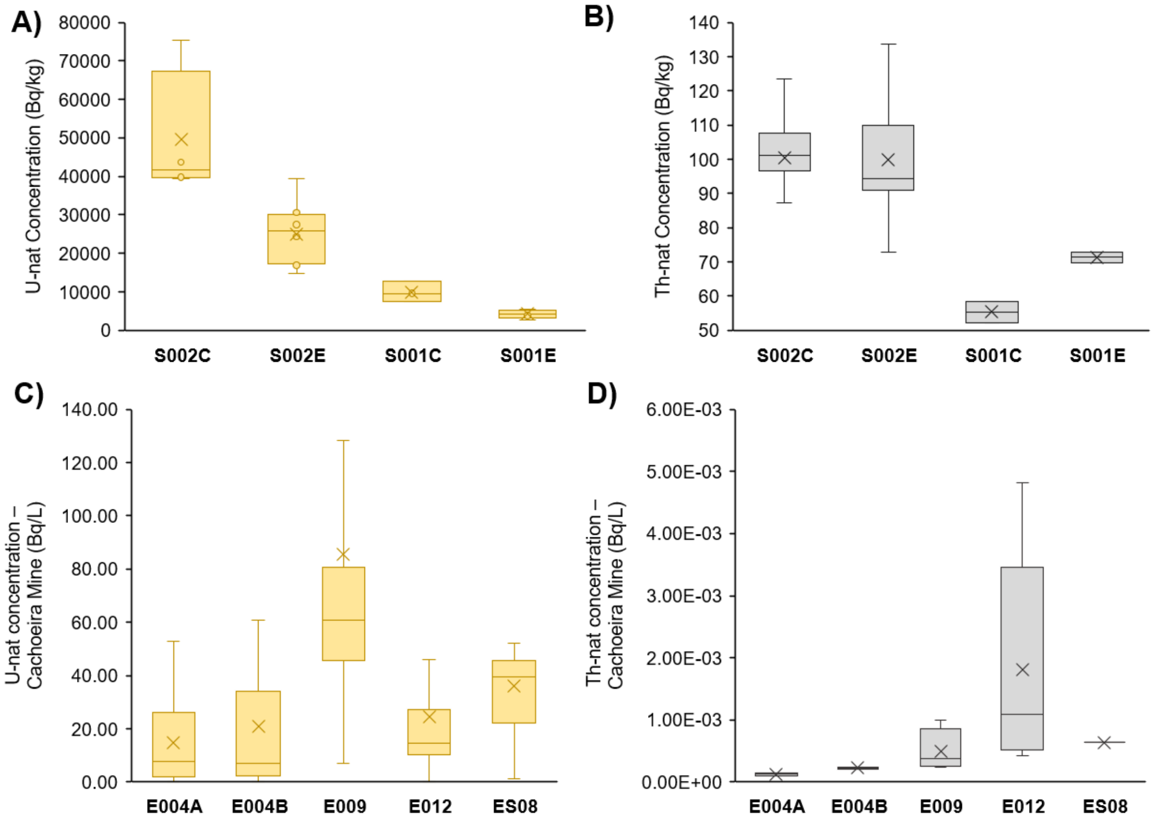
This finding aligns with the historical ore extraction patterns of the mining company, which initially removed large amounts of high-grade ore from the Cachoeira mine, in accordance with the amount currently extracted from the Engenho mine [49]. Since Engenho contains lowergrade ore, a larger volume must be processed to obtain the same amount of uranium, leading to increased waste generation and a potentially greater environmental impact compared to Cachoeira.

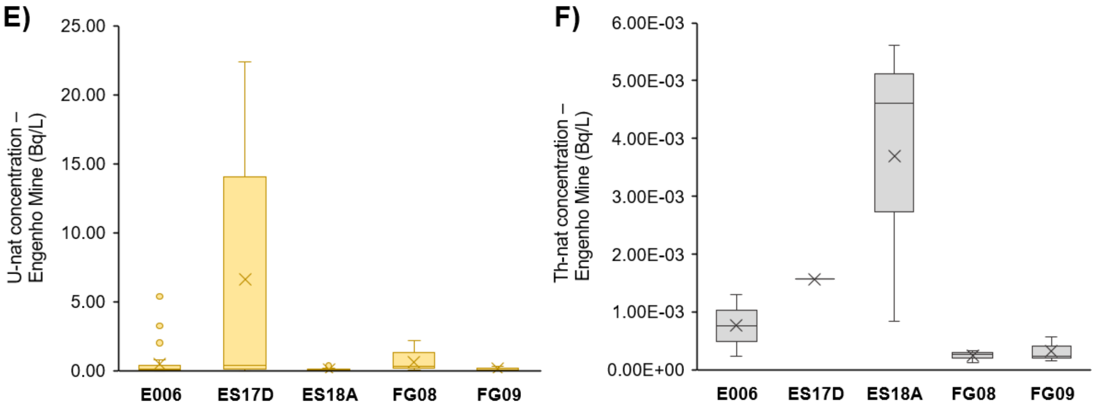
As discussed by Porto et al. (2024) [49], the company's uranium production profile has evolved over the years, largely influenced by the radiochemical characteristics of the mines. Production initially relied on the Cachoeira mine, but after a period of inactivity, operations resumed with ore from the Engenho mine. The variation in uranium levels between these mines reflects differences in their geological formations, showing that, while the region as a whole has a high uranium background, not all areas within the mining zone exhibit the same radiochemical profile [50].

It is known from the literature that the concentration of uranium in uranium ore varies widely depending on the source and type of ore. A study reported uranium concentrations as high as 2600 ± 100 Bq.kg^{-1} in samples from the Mezdreja uranium mine tailing dump, in Serbia [51]. Similarly, another study found maximum uranium concentrations of 81.5 mg.kg^{-1} (2070 Bq.kg^{-1}) in stream sediments downstream of an open pit lake and dumps [52].

Both the Cachoeira and Engenho mines exhibit uranium concentrations that are higher than those reported in the literature, suggesting that the uranium levels in this region are notably elevated, even when compared to other uranium mining sites.

Fig. 1. U-nat and Th-nat concentrations in ores and aqueous samples in both of the mines. For A and B, the sample name endings indicate their origin: C – Cachoeira and E – Engenho. A) U-nat concentration of ore samples (Bq/kg). B) Th-nat concentration of ore samples (Bq/kg). C) U-nat concentration of aqueous samples in the Cachoeira Mine (Bq/L). D) Th-nat concentration of aqueous samples in the Cachoeira Mine (Bq/L). mines. E) U-nat concentration of aqueous samples in the Engenho Mine (Bq/L). F) Th-nat concentration of aqueous samples in the Engenho Mine (Bq/L).





Regarding radiological impact, the literature shows that uranium ores contain uranium with a natural isotopic composition in concentrations ranging from less than 0.03% to more than 20% [53]. As stated earlier, in uranium ore deposits, secular equilibrium occurs between U-238 and its decay products, and between U-235 and its decay products, respectively [48]. This balance can be slightly disturbed by geochemical migration processes in the ore deposit [54].

In the case of an undisturbed uranium deposit, which is not the case of the mines studied here, the activity of all decay products remains constant for hundreds of millions of years [55]. In the ore deposit, radiation is practically trapped underground. Radon is not a concern for deep deposits, although it can travel through underground fissures as it decays before reaching the surface [56]. The situation changes completely when the deposit is mined: radon gas can escape into the air, ore dust can be dispersed by wind, and contaminants can leach and infiltrate surface water bodies and groundwater [57], which is the case of these mines.

However, the mining operations at these sites are carried out through open-pit mining, which significantly reduces the concentration of radon in the surrounding areas. Open-pit mining exposes the ore directly to the atmosphere, allowing radon gas to dissipate quickly rather than accumulate in underground spaces. While this reduces radon-related risks in the vicinity of the mining site, other radionuclides present in the ore can still contribute to environmental radiation exposure, making continuous monitoring essential to ensure the safety of workers and nearby communities [58].

3.1.2. Natural Thorium

Samples of crushed ores from the Cachoeira and Engenho mines were also analyzed for their natural thorium content (Figure 1B). Despite presenting a discrepant statistical dispersion, the natural thorium values for the ores from each of the mines present very similar averages, which could allow a preliminary conclusion that the radiochemical profile referring to Th-232 in both mines is quite similar. The crushed ore from Cachoeira (S002C) has 100.37 Bq/kg, while Engenho's crushed ore (S002E) has 99.98 Bq/kg, which are nearly identical average values. This suggests that the thorium content in both mines is relatively comparable, showing that both deposits contain similar amounts of thorium in their ores.

When examining the leachate, however, there is a noticeable difference. Cachoeira's leachate (S001C) contains 55.40 Bq/kg, while Engenho's leachate (S001E) has 71.32 Bq/kg. This trend indicates that more thorium is retained in the leachate from Engenho, which may suggest that the extraction process is less efficient in removing thorium from the ore in this mine, or that the thorium in Engenho is more easily leached.

Several factors may account for the differences observed in thorium levels between the leachates. In the case of the Engenho ore, thorium might be linked to mineral phases that are more prone to dissolution or more weakly bound within the solid matrix, which would facilitate its release during leaching. Furthermore, physical characteristics such as particle size distribution, pore structure, and the presence of accessory minerals like clays or iron oxides could significantly impact thorium behavior during processing. Also, minor differences in the geochemical environment (shifts in pH, oxidation-reduction potential, or the availability of complexing ligands like organic compounds or phosphate ions) may enhance thorium solubility and mobility in the Engenho samples. It is known that the presence of sulfate, phosphate, and organic ligands (e.g., fulvic acid) can form stable thorium complexes, greatly enhancing its solubility and mobility, especially under acidic to neutral conditions [59–61].

Another key aspect is the variability in the data, as indicated by the standard deviations. Cachoeira's crushed ore (S002C) shows a standard deviation of 17.38 Bq/kg, while Engenho's crushed ore (S002E) has a slightly higher standard deviation of 17.77 Bq/kg. These relatively close values suggest similar levels of variability in the thorium distribution in both mines. However, when we look at the leachate, Cachoeira (S001C) has a much lower standard deviation of 3.08 Bq/kg, indicating less fluctuation in the thorium content in the leachate compared to Engenho, which has a higher standard deviation of 1.58 Bq/kg. This points to the fact that the thorium content in the leachate from the Engenho mine is more uniform and possibly more influenced by the composition of the ore being processed.

When evaluating the efficiency of thorium extraction, the ratio between the thorium concentration in the leachate (S001) and crushed ore (S002) can provide some insights. For Cachoeira mine, this ratio is 0.5538, meaning that approximately 55.40% of the thorium from the crushed ore is transferred to the leachate. In contrast, Engenho has a ratio of 0.7146, indicating that about 71.50% of the thorium in the crushed ore is transferred to the leachate. This higher ratio in Engenho may confirm that thorium is more readily leached from the ore in this mine, as discussed previously.

The literature shows that in the province of Gauteng in South Africa, average activity concentrations of Th-232 in mine tailings are 43.9 ± 1.0 $Bq.kg^{-1}$ [62]. Similarly, in the tin mining area of Jos Plateau, Nigeria, the activity concentrations of thorium-232 ranged from 5,290 to 47,500 $Bq.kg^{-1}$ [63]. The average concentration of thorium in Malaysian monazite ore, in turn, was found to be 17,990.5 ± 1,239.3 ppm (73,041 ± 5,031.6 $kBq.kg^{-1}$) [64].

In contrast to the Gauteng mining tailings, the concentrations in the Brazilian mines (Cachoeira and Engenho) are significantly greater, suggesting that the thorium levels in these particular Brazilian mines may be higher. Nonetheless, the range of concentrations is similar to that of the Jos Plateau tin mining zone, indicating a comparable thorium content in these Brazilian mines and the particular Nigerian mining location. Nevertheless, these concentrations remain far below the extremely high levels found in Malaysian monazite ore, which exhibits significantly greater radioactivity.

It was expected that the natural thorium concentrations in the Brazilian Cachoeira and Engenho mines would be relatively low compared to other mining regions, given that these mines are focused on uranium extraction, not on thorium.

3.2 Aqueous Samples

3.2.1 Natural Uranium

Figures 1C and 1E show a boxplot for the natural uranium values (in $Bq.L^{-1}$) of samples collected around and inside the Cachoeira and Engenho mines, respectively. Rainwater that runs over the waste and leached ore deposit and liquids from the Cachoeira mine are received by E004A. The pre-basin overflows into E004B, and the uranium content there is somewhat greater than in E004A. This would suggest that the fines sedimentation basin is perhaps showing a subtle uranium leakage.

When compared to the other collection points, the uranium level at E009, which is situated inside mine pit 1, is noticeably greater. This was already expected, once there is a comparatively high concentration of uranium in the mining pit [65–67]. While not as high as it was at E009, the uranium level in E012 is still rather significant. ES08, in turn, receives water originating from the pumping of water accumulated in Cachoeira Mine pits, which then flows over the piles of the Waste and Leached Ore Deposit, also showing some level of uranium content.

At the fines sedimentation basin (E006), the uranium content is rather low. ES17D (mine pit II) has much more uranium than the fines sedimentation basin (E006). In ES18A, uranium content is comparatively low. Nonetheless, there may be differences in certain areas in mine pit III. The comparatively low uranium level in FG08 suggests that there is little uranium content in the geological exploratory drilling holes. Additionally, FG09's uranium content is quite low, as also seen in FG08.

As shown in Figure 1A, the Cachoeira Mine exhibits average uranium concentrations more than twice as high as those at the Engenho Mine at certain points, indicating a considerable overall difference in uranium levels between the two sites. This confirms how different the radiochemical profile is compared between the mines and reinforces the heterogeneity of the uranium distribution due to the anomalies explored by the mining company.

It is well established that the uranium concentration in seawater is very low, at only 3.3 ppb (0.084 Bq.L^{-1}), due to its low solubility [68]. In contrast, uranium concentrations in carbonate ore leach solutions can be much higher, often 1 g.L^{-1} (25,400 Bq.L^{-1}) [69]. The equilibrium uranium concentration in water resulting from the interaction with rocks or uranium ore is only $\leq 10^{-8}$ mol.L^{-1} ($\leq$ 0.604 Bq.L^{-1}) [70].

Studies have shown that uranium concentrations in water samples from mining areas can exceed recommended guidelines, leading to enhanced radioactivity levels in surface water streams [71]. The solubility processes of uranium under oxidizing conditions in soils partially saturated with water can lead to concentrations in residue pore fluids that exceed solubility limits for U(VI)-containing [72]. This is further supported by findings that uranium concentrations in borehole and shallow well water samples ranged from 7 to 263 µg.L^{-1} (0.18 to 6.68 Bq.L^{-1}) and 17 to 199 µg.L^{-1} (0.43 to 5.1 Bq.L^{-1}), respectively [73].

The impact of uranium mining on water quality is also evident in studies that have reported elevated uranium levels in lake water, waste heaps, adjacent soils, and groundwater in mining regions [74]. A study investigating the impact of a disused mine on uranium transport in a river in South West England further highlights the potential for uranium contamination in water bodies [75]. Values of 1268 µg.L^{-1} (32.20 Bq.L^{-1}) in open pit lakes, 100 µg.L^{-1} (2.54 Bq.L^{-1}) in surface water, 103 µg.L^{-1} (2.61 Bq.L^{-1}) in groundwater [52] were found in a study that analyzed uranium in a stream downstream of an open pit lake and dumps [52].

When comparing the estimated uranium concentrations in the Engenho and Cachoeira mines with those found in the literature, it becomes clear that both mines exhibit significantly higher uranium levels in water. The Cachoeira mine, for example, with an average estimated concentration of approximately 36.3 Bq.L^{-1}, appears to be more similar to the values mentioned in the literature than the Engenho mine, with an estimated concentration of approximately 1.8 Bq.L^{-1}.

The examples provided in the literature, such as seawater, carbonate ore leach solutions, and rock interactions, represent different types of water samples that differ significantly from those collected in the Engenho and Cachoeira mines. These differences are largely due to the specific geological and hydrological contexts in which the uranium ores are located. Interactions between uranium ores and the surrounding rock formations in different geological environments significantly influence how readily uranium can dissolve in water.

3.2.2 Natural Thorium

Figures 1D and 1F show a boxplot for the natural thorium values (in Bq.L^{-1}) of samples collected around and inside the Cachoeira and Engenho mines. Significant differences in the amounts of this natural thorium can be seen in different parts of the mine based on natural thorium data that was gathered at several locations inside Cachoeira Mine.

Locations E004A and E004B, which are situated in the fines sedimentation basin and the pre-basin, respectively, show natural thorium levels that are rather low and stable. This might indicate that the streams passing through these regions have a consistent concentration of thorium in them. It was also observed a notable rise in natural thorium levels upon entering the mine at point E009, which is situated within Cachoeira mine's pit 1, which was already expected, as it refers to an internal point within the mine.

In pit 2 of the mine, at position E012, thorium levels are considerably higher, which implies that pit 2 could have some little thorium geological anomalies. Lastly, natural thorium at point ES08, which is in the interception channel at the base of the waste rock pile, are comparable to those at pit 2 (E012). This implies that significant amounts of thorium may be transported by pumping water out of the pits [76].

In general, the natural thorium data collected at the Engenho mine showed a significant variation in the element's concentrations at different points in the mine. It can be observed that natural thorium values are rather low, starting with locations FG08 and FG09, which correspond to geotechnical drilling holes at the Engenho mine. On the other hand, natural thorium concentrations are significantly greater at locations ES17D and ES18A, which are situated in the in-pit regions of pits II and III of the mine. These locations are connected to mine drainage and have increased direct exposure to uranium ore.

In addition, thorium levels at Point E006, which is associated with the Engenho mine fines sedimentation basin, are comparatively greater than those at geotechnical drilling holes. This implies that there could be significant amounts of naturally occurring thorium in the waters that flow and collect in this area.

A study reported even higher thorium concentrations of up to 38 ppm (154.28 Bq.L^{-1}) in seeps and ground waters associated with uranium mining in the United States and Canada [77]. Another research reported mean activity concentrations of thorium in water ranging from 0.002 to 0.037 Bq.L^{-1} in the Oum Er-Rabia river, indicating the presence of thorium in natural water bodies affected by uranium mining [78].

The range of thorium levels in various samples in Cachoeira mine is 1.27 x 10^{4} to 1.8 x 10^{3} Bq.L^{-1}, but the range at the Engenho mine is 2.41 x 10^{4} to 3.69 x 10^{3} Bq.L^{-1}. A significant discrepancy is seen when these results are compared to previous research, which showed results of more than 150 Bq.L^{-1}. The disparity in thorium concentrations between Brazilian mines and the locations reported in the literature is highlighted by this significant variance. Although the concentrations in the literature samples are much greater than those in the Engenho and Cachoeira mines, this suggests that distinct topographical and operational factors exist in each environment.

Furthermore, the radionuclide with the highest known abundance in the mines analyzed in this study is uranium. In this sense, in fact, it was not expected that thorium concentrations, although relatively significant, could be higher than those of average ore samples from other mines reported in the literature.

3.3 Soil Samples

3.3.1 Natural Uranium

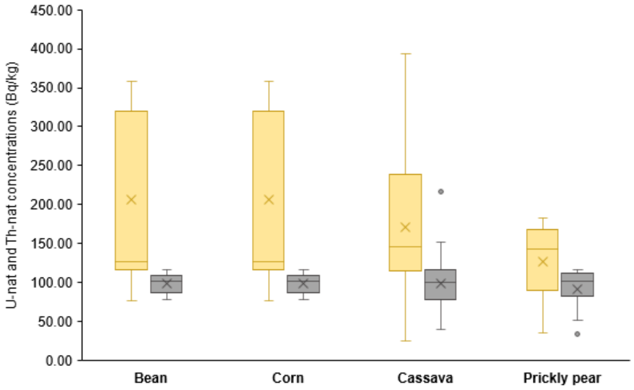
This study's soil samples originate from isolated areas near the mines, where crops and cattle are raised. These soils are essential for assessing the amounts of uranium in agricultural and animal products, as well as for comparing the uranium content of mine ores.

Uranium may seep into the soil from adjacent geological formations, influencing the surrounding ecosystem and perhaps changing the makeup of crops and fodder [79]. Soil contamination by uranium can lead to a significant change in the fungal community, affecting the soil ecosystem under experimental conditions [80].

The mobility and bioavailability of uranium in soils are influenced by various factors such as pH, soil type, and physicochemical properties [81,82]. The sorption of uranium in soils is affected by pH, with significant impacts on the total and available uranium content in the soil [83]. In this sense, uranium-contaminated soils can also lead to uranium entering the human food chain through water and plants, posing a threat to human health and the environment [84]. The uptake of uranium by crops, such as maize and wheat, is influenced by the soil type, with uranium concentration in the soil decreasing with an increase in soil acidity, contrasting its accumulation in the roots of cultivated plants [85].

Figure 2 shows the concentration of natural uranium and thorium in Bq.kg^{-1} in soils associated with bean, corn, cassava and prickly pear plantations.

Fig. 2. U-nat (yellow) and Th-nat (gray) results of soil samples.



There is a discernible variation in the amounts of uranium present in the various farming soils. Beans and corn soils share the same uranium content, which is higher than that of cassava and prickly pear ones. The disparities in uranium content may be attributed to a combination of environmental factors and also perhaps agricultural practices, but with a minimum contribution. Factors like soil composition, water sources, and land use can influence uranium levels. It is notable that the difference of approximately 1000 orders of magnitude between the soils used as reference and the samples from the mines can allow a reliable comparison, whose reference values are, in fact, the background values of the region.

Several studies have investigated the behavior of uranium in soil and its potential extraction and remediation methods. Research demonstrated that the residual concentration of uranium-citrate complexes in treated soils can be rapidly reduced by maintaining the soil water pH between 8 and 9 [86]. Additionally, another study highlighted the influence of uranium per unit surface area of soil particles, sorption site variability, and soil mineralogy on the reaction rate during carbonate leaching of uranium from contaminated soils [87]. Furthermore, another study reported specific uranium concentrations in soil samples from former uranium mining sites in Germany, highlighting the existing body of literature focused on uranium determination in soil matrices [88].

Some studies presented the detection of high levels of uranium in soil and groundwater samples, with concentrations reaching 350 $\mu g.g^{-1}$ (8,890 $Bq.kg^{-1}$) and varying from 5 $ng.g^{-1}$ to 2 $\mu g.g^{-1}$ (0.12 to 50.8 $Bq.kg^{-1}$) in soil layers and water, respectively [89,90].

The values found in agricultural crop soils (Bean, Corn, Cassava, Prickly pear) are significantly lower than the values mentioned in the literature for soil (350 $\mu g.g^{-1}$ or 8,890 $Bq.kg^{-1}$). Agricultural crops have concentrations that are in the range of tens of $Bq.kg^{-1}$, while literature values are much higher. This shows that, although the agricultural products are collected from a region that is relatively close to the mines, the distance to it of at least 20 km shows that the contribution of the mines may be minimal or even none at all. In this sense, it can be inferred that the uranium concentration in the soil is the background of the region itself.

It is important to highlight that the concentrations observed in some agricultural soils (50–400 $Bq \cdot kg^{-1}$ for U) may appear elevated compared to global natural averages. However, the region is characterized by known geochemical anomalies, which explains part of this variability. To contextualize the radiological impact, we performed a preliminary dose estimation using IAEA conversion factors. The results indicate that, even at the highest values measured, the annual effective dose for external exposure remains below the reference public limit of 1 $mSv \cdot y^{-1}$. Nevertheless, we recommend that future studies include detailed radiological risk modeling and continuous monitoring to better evaluate potential health risks.

3.3.2 Natural Thorium

The soil samples analyzed here were the same soils analyzed for uranium, previously shown (Figure 2). These soils are very important for evaluating the thorium concentration of mine ores and for determining the thorium content of agricultural and animal products.

The levels of natural thorium in soils from both crops show a smaller fluctuation when compared to the variation between natural uranium values in the same soils. In this sense, it is possible that the region's natural thorium background may, for some locations, be relatively more homogeneous and well distributed than the uranium profile.

There is a study in the literature that reported average activity concentrations of Th-232 in non-affected areas of uranium mining to be 28 ± 39 Bq.kg^{-1} [91]. Another research observed activity concentrations of thorium in affected areas ranging from 1.57 to 26.9 mg.kg^{-1} (6.37 to 109.21 Bq.kg^{-1}) [92]. Soil around the Long Term Storage Facility in Bukit Kledang, Perak, Malaysia, showed a radioactivity concentration of 20.83 ± 5.88 Bq.kg^{-1} [93].

The results show the amounts of thorium activity in crops soil used for agriculture, ranging from 90.97 to 99.42 Bq.kg^{-1}. On the other hand, typical Th-232 concentrations in non-uranium mining impacted areas are reported in the literature to be around 28 ± 39 Bq.kg^{-1}. This discrepancy shows that crop soil thorium concentrations are greater than those found in unaffected areas. In regions impacted by uranium mining, thorium concentrations were found to range from 6.37 to 109.21 Bq.kg^{-1}, according to another study published in the literature. It is notable that the values in the crops' soils are comparable to or greater than the concentrations discovered in regions affected by mining when these data are compared with the concentrations in agricultural crops soils.

Moreover, the radioactivity content in the soil around the long-term storage facility at Bukit Kledang, Perak, Malaysia, was 20.83 ± 5.88 Bq.kg^{-1}, which is similar to what is seen in crops soils. According to this investigation, thorium concentrations in soils of agricultural products are significant when compared to levels reported in areas unaffected by uranium mining, and in certain instances, they are even greater than those found in places where mining has a direct influence. In general, the thorium concentrations measured in these agricultural soils are consistent with the values reported in the literature for both unaffected and uranium mining-impacted areas.

3.4 COMPARISONS

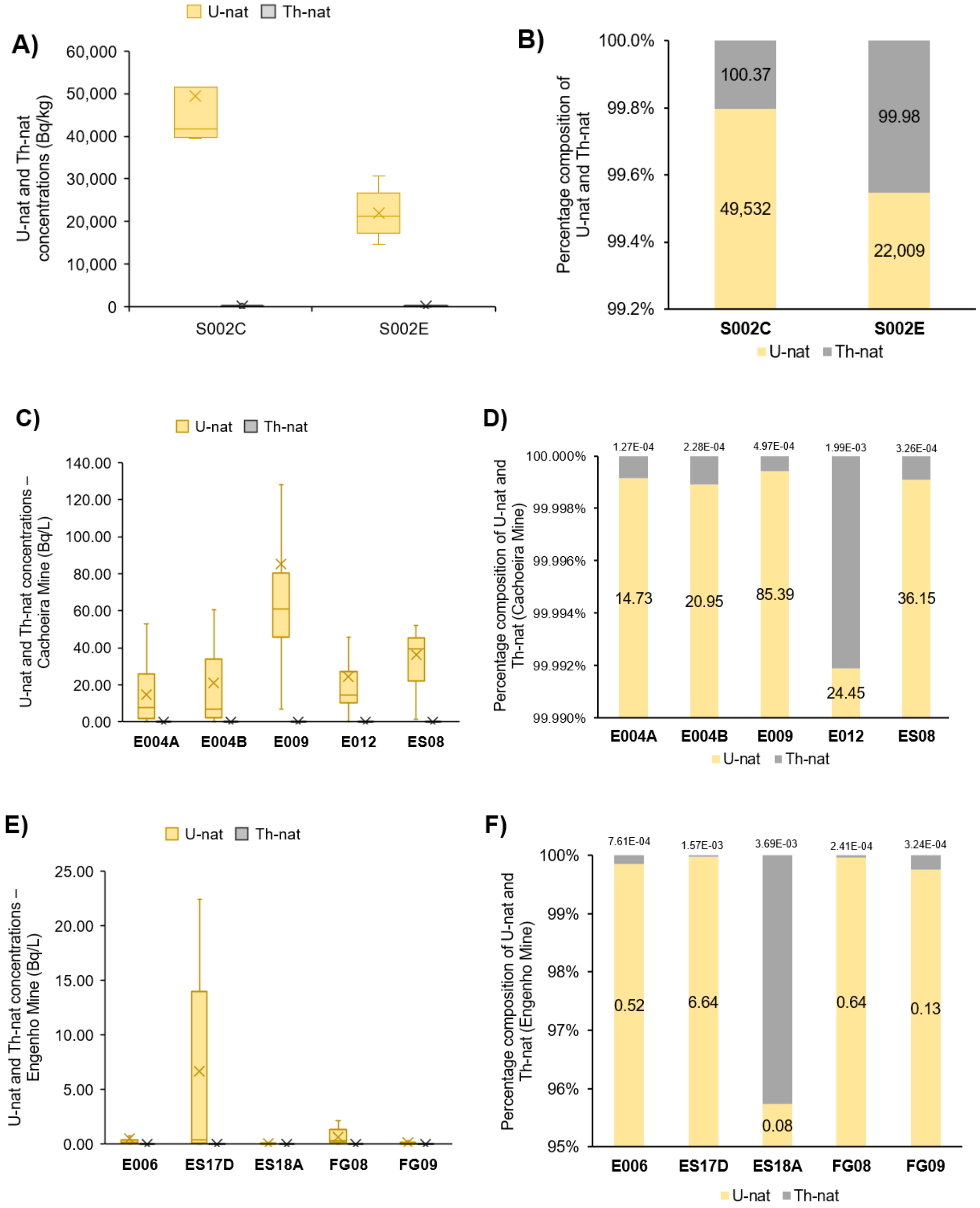
In this section, we integrate the results obtained for different matrices (ores, waters, and soils) and highlight radiogeochemical patterns between the Cachoeira and Engenho mines. The purpose is to help the reader understand not only the absolute values, but also the contrasts between U-nat and Th-nat and their different environmental mobilities.

3.4.1 Ore Samples

Despite the knowledge that the uranium ore types from the Cachoeira and Engenho mines exhibit distinct radiochemical signatures, this topic will undertake a comparative analysis between them. Figures 3A shows a comparative boxplot with ore values from both mines.

Based on the analysis of the boxplot referring to the ores from the Cachoeira and Engenho mines, it can be seen that the discrepancy between the values of natural uranium and natural bull are very significant, which shows that in fact the ores have a great abundance of uranium when compared to thorium. In this sense, to carry out a more quantitative analysis regarding the radiochemical profile of U-nat and Th-nat from both mines, the aqueous samples from each of the Cachoeira mines and the Engenho mine will be analyzed comparatively.

Fig. 3. A) U-nat and Th-nat results of uranium ores from Cachoeira Mine (end C) and Engenho Mine (end E). B) Percentage composition of U-nat and Th-nat in ore samples from both mines. Label values are concentrations in Bq.kg^{-1}. C) U-nat and Th-nat results of aqueous samples from Cachoeira Mine. D) Percentage composition of U-nat and Th-nat in aqueous samples from Cachoeira Mine. Label values are concentrations in Bq.L^{-1}. E) U-nat and Th-nat results of aqueous samples from Engenho Mine. F) Percentage composition of U-nat and Th-nat in aqueous samples from Engenho Mine. Label values are concentrations in Bq.L^{-1}.



Using the averages to make a proportional comparison between the U-nat and Th-nat values of each mine (Figure 3B), it can be seen that the radiochemical profile of natural uranium is almost 500 times higher than that of natural thorium in the Cachoeira mine. On the other hand, at the Engenho mine, this proportion reaches around 220. This means that, in fact, the composition of the ore at the Engenho mine has a higher natural thorium content than the Cachoeira mine, being almost twice higher in percentage terms.

3.4.2 Aqueous Samples

Naturally, just like the U-nat and Th-nat values in the ores from each mine, the liquid samples collected from each mine also, it is expected, may present very different radiochemical profiles. This is an analysis that will be done in this topic. Figure 3C shows a boxplot of U-nat and Th-nat results of aqueous samples from Cachoeira Mine.

Analyzing the radiochemical profile of the aqueous samples from Cachoeira Mine, it can be seen that the discrepancy between the values of natural uranium and natural torus is still very large. This is an expected result, since the nature of the ore directly influences the nature of the soil and, consequently, the water that is eventually represented in and around the mine.

To make a more effective comparison between natural uranium and thorium in the Cachoeira mine aqueous samples possible, the percentage composition axis was adapted to the third decimal place, going from 99.990% to 100.000%. This shows that, in fact, the abundance of uranium relative to thorium is very significant.

It is important to highlight that the percentage composition analysis does not imply that the components of the aqueous samples are only natural uranium and thorium. This representation actually shows the main radionuclides that lead a nuclear decay series: U-238 and Th-232. The concentration of the radionuclides of each series is expected to be approximately in secular equilibrium with its parent nuclide. In other words, this means that the concentrations of Ra-226 and Pb-210, radionuclides with a relatively long half-life, present subtly similar concentrations. Likewise, Ra-228, the immediate decay son of Th-232, should present relatively close concentrations (or possibly in the same order of magnitude) as its parent. Figure 3E shows U-nat and Th-nat concentrations in aqueous samples from Engenho Mine.

Similarly regarding the results of aqueous samples from the Cachoeira mine regarding U-nat and Th-nat, the values are in tens of orders of magnitude of difference. In this sense, a more assertive analysis will be carried out based on the percentage composition profile of these radionuclides.

Figure 3F shows the percentage composition of U-nat and Th-nat in aqueous samples from Engenho Mine. Unlike the representation referring to Cachoeira mine, it was not necessary to use the third decimal place to evaluate the difference in the profile of these radionuclides.

The percentage composition referring to the aqueous samples from the Engenho mine shows a trend already observed in the comparative analysis of both ore samples from the two mines. The proportional quantity of thorium relative to uranium is higher in the Engenho mine, a profile that was present not only in the ores, but naturally, in the aqueous samples.

3.4.3 Soil Samples

Also used as reference samples for values in the region around the mines, soils are one of the matrices that already present similar orders of magnitude for natural uranium and thorium.

Despite presenting discrepancies when comparing the results of uranium and natural thorium with each other for each one, the soils present a more constant behavior when compared to the other matrices previously presented. Furthermore, it is noted that thorium concentrations in different soils show a linearity that is not observed in uranium, which confirms the hypothesis that thorium is found in a more dispersed and homogeneous form in the surroundings of uranium mining.

3.5. Limitations and Perspectives

This study has some limitations: (i) the number of sampling points, although representative, does not cover the entire area of influence of the mines; (ii) the lack of temporal series prevents assessment of seasonal variations in U and Th mobility; (iii) a complete radiological risk model was not performed.

Future research should include continuous monitoring of surface and groundwater, evaluation of agricultural products and biological matrices, and application of radiological dispersion models.

From a broader perspective, the findings presented here are not restricted to the Cachoeira and Engenho mines but provide insights for sustainable uranium mining management in other regions, reinforcing the need for public policies aimed at environmental protection and community health.

Conclusions

In this study, we effectively employed ICP-MS to quantify naturally occurring uranium and thorium across a variety of environmental matrices, including ores, water samples, and soils, providing insights into how these radionuclides are distributed in areas influenced by uranium mining.

Clear differences in uranium concentrations were observed between the Cachoeira and Engenho mines, reflecting the distinct radiochemical signatures of each deposit. Uranium levels at the Cachoeira mine were found to be significantly higher than thorium, with a uranium-to-thorium ratio nearly twice that of the Engenho mine. While results from Engenho are consistent with values commonly reported in the literature, the elevated concentrations at Cachoeira underscore the uniqueness of its ore composition.

This trend was also evident in the aqueous samples, where uranium remained consistently more abundant than thorium, especially in samples from Cachoeira. At both sites, uranium appears to be more mobile, which may explain its pronounced presence in the water.

Soil samples presented a somewhat different profile. Although variations were still observed, uranium and thorium concentrations generally fell within the same order of magnitude. Notably, thorium showed a more uniform distribution across the sampling sites, likely due to its lower solubility and greater affinity for soil particles.

The values we obtained align well with those reported in the literature and fall within the expected range for such matrices around a uranium mining site. These results emphasize the role of local geological and geochemical factors in shaping the environmental behavior of uranium and thorium.

In summary, the results confirm that U and Th concentrations in the region reflect both the natural geological background and the influence of mining activities. The higher mobility of uranium compared to thorium indicates a greater potential for transport in water, which should be carefully considered in monitoring programs. By examining multiple sample types, we gained a more comprehensive understanding of how these radionuclides are distributed and transported, an important step toward improving environmental monitoring in areas close to uranium mines.

Author Contributions Statement

Joel Augusto Moura Porto: Investigation; methodology; writing – original draft; software; data curation; formal analysis; visualization; writing – review and editing; software. **Lander de Jesus Alves:** Investigation; writing – original draft; formal analysis; visualization; writing – review and editing. **Hector Hugo Silva Medrado:** Investigation; methodology; visualization; writing – review and editing; software. **Josilene da Silva Rocha:** Investigation; formal analysis; visualization; writing – review and editing. **Rodrigo Gibaut de Souza Góis:**

Investigation; formal analysis; visualization; writing – review and editing. **Fábio Carvalho Nunes**: Investigation; formal analysis; writing – review and editing. **Marcelo Machado Viana:** Conceptualization; investigation; writing–review and editing; supervision; project administration; methodology; resources. **Ana Paula de Carvalho Teixeira:** Conceptualization; investigation; writing–review and editing; supervision; project administration; methodology; resources.

Conflict of Interest Statement

There are no conflicts to declare.

Data Availability Statement

Todo o conjunto de dados de apoio aos resultados deste estudo foi disponibilizado em Science DataBase e pode ser acessado em https://www.scidb.cn/en/s/7Nj6vu

References

[1] J. Fang, G. Gozgor, M.K. Mahalik, H. Padhan, R. Xu, The impact of economic complexity on energy demand in OECD countries, Environ. Sci. Pollut. Res. 28 (2021) 33771–33780. https://doi.org/10.1007/s11356-020-12089-w.
[2] T. Delany-Crowe, D. Marinova, M. Fisher, M. McGreevy, F. Baum, Australian policies on water management and climate change: are they supporting the sustainable development goals and improved health and well-being?, Glob. Health 15 (2019) 68. https://doi.org/10.1186/s12992-019-0509-3.
[3] T.F. Agbanike, C. Nwani, U.I. Uwazie, L.I. Anochiwa, T.-G.C. Onoja, I.O. Ogbonnaya, Oil price, energy consumption and carbon dioxide (CO2) emissions: insight into sustainability challenges in Venezuela, Lat. Am. Econ. Rev. 28 (2019) 8. https://doi.org/10.1186/s40503-019-0070-8.
[4] J.A. Fuinhas, M. Koengkan, N.C. Leitão, C. Nwani, G. Uzuner, F. Dehdar, S. Relva, D. Peyerl, Effect of Battery Electric Vehicles on Greenhouse Gas Emissions in 29 European Union Countries, Sustainability 13 (2021) 13611. https://doi.org/10.3390/su132413611.
[5] F. Guerhardt, T.A.F. Silva, F.M.C. Gamarra, S.E.R. Ribeiro Júnior, S.A.V. Llanos, A.P.B. Quispe, M. Vieira Junior, E.B. Tambourgi, J.C.C. Santana, R. Maria Vanalle, A Smart Grid System for Reducing Energy Consumption and Energy Cost in Buildings in São Paulo, Brazil, Energies 13 (2020) 3874. https://doi.org/10.3390/en13153874.
[6] B. Muhammad, M.K. Khan, M.I. Khan, S. Khan, Impact of foreign direct investment, natural resources, renewable energy consumption, and economic growth on environmental degradation: evidence from BRICS, developing, developed and global countries, Environ. Sci. Pollut. Res. 28 (2021) 21789–21798. https://doi.org/10.1007/s11356-020-12084-1.
[7] V.P. Guinn, Radioactivity, in: Encycl. Phys. Sci. Technol., Elsevier, 2003: pp. 661–674. https://doi.org/10.1016/B0-12-227410-5/00643-8.
[8] E. González, J.M. Martínez-Val, Nuclear Energy: World Perspectives, in: A. Marquina (Ed.), Energy Secur., Palgrave Macmillan UK, London, 2008: pp. 232–243. https://doi.org/10.1057/9780230595002_15.

[9] J. Iqbal, F. M. Howari, A.-M.O. Mohamed, E.K. Paleologos, Assessment of radiation pollution from nuclear power plants, in: Pollut. Assess. Sustain. Pract. Appl. Sci. Eng., Elsevier, 2021: pp. 1027–1053. https://doi.org/10.1016/B978-0-12-809582-9.00020-7.

[10] R. Ali, T.M.N.T. Mansur, N.H. Baharudin, S.I.S. Hassan, Environmental impacts of renewable energy, in: Electr. Renew. Energy Syst., Elsevier, 2016: pp. 519–546. https://doi.org/10.1016/B978-0-12-804448-3.00021-9.

[11] M. Lehtveer, F. Hedenus, Nuclear power as a climate mitigation strategy – technology and proliferation risk, J. Risk Res. 18 (2015) 273–290. https://doi.org/10.1080/13669877.2014.889194.

[12] M. Cooper, Nuclear safety and affordable reactors: Can we have both?, Bull. At. Sci. 68 (2012) 61–72. https://doi.org/10.1177/0096340212451627.

[13] H. Zhang, J.-P. Li, W.-L. Chen, E.-B. Wang, Application of sandwich-type polyoxometalates modified TiO<sub>2</sub> in dye-sensitized solar cells, Chin. Sci. Bull. 63 (2018) 3333–3341. https://doi.org/10.1360/N972018-00632.

[14] M.A.J. Huijbregts, L.J.A. Rombouts, S. Hellweg, R. Frischknecht, A.J. Hendriks, D. Van De Meent, A.M.J. Ragas, L. Reijnders, J. Struijs, Is Cumulative Fossil Energy Demand a Useful Indicator for the Environmental Performance of Products?, Environ. Sci. Technol. 40 (2006) 641–648. https://doi.org/10.1021/es051689g.

[15] U. Chakravorty, B. Magne, M. Moreaux, Resource Use under Climate Stabilization: Can Nuclear Power Provide Clean Energy?, J. Public Econ. Theory 14 (2012) 349–389. https://doi.org/10.1111/j.1467-9779.2011.01545.x.

[16] F. Adler, H. Von Halban, Nuclear Physics: Control of the Chain Reaction involved in Fission of the Uranium Nucleus, Nature 143 (1939) 793–793. https://doi.org/10.1038/143793a0.

[17] A.C. Miller, ed., Depleted Uranium: Properties, Uses, and Health Consequences, 1st ed., CRC Press, 2006. https://doi.org/10.1201/9781420004564.

[18] R. Prăvălie, G. Bandoc, Nuclear energy: Between global electricity demand, worldwide decarbonisation imperativeness, and planetary environmental implications, J. Environ. Manage. 209 (2018) 81–92. https://doi.org/10.1016/j.jenvman.2017.12.043.

[19] F. Winde, D. Brugge, A. Nidecker, U. Ruegg, Uranium from Africa – An overview on past and current mining activities: Re-appraising associated risks and chances in a global context, J. Afr. Earth Sci. 129 (2017) 759–778. https://doi.org/10.1016/j.jafrearsci.2016.12.004.

[20] L.S. Gottlieb, L.A. Husen, Lung Cancer Among Navajo Uranium Miners, Chest 81 (1982) 449–452. https://doi.org/10.1378/chest.81.4.449.

[21] L. Tomásek, S.C. Darby, Recent results from the study of West Bohemian uranium miners exposed to radon and its progeny., Environ. Health Perspect. 103 (1995) 55–57. https://doi.org/10.1289/ehp.103-1518834.

[22] S. Cornacchia, R. Errico, L. La Tegola, A. Maldera, G. Simeone, V. Fusco, A. Niccoli-Asabella, G. Rubini, G. Guglielmi, The new lens dose limit: implication for occupational radiation protection, Radiol. Med. (Torino) 124 (2019) 728–735. https://doi.org/10.1007/s11547-019-01027-7.

[23] K. Moeng, Community perceptions on the health risks of acid mine drainage: the environmental justice struggles of communities near mining fields, Environ. Dev. Sustain. 21 (2019) 2619–2640. https://doi.org/10.1007/s10668-018-0149-4.

[24] J.D. Eakins, The application of radiochemical separation procedures to environmental and biological materials, Nucl. Instrum. Methods Phys. Res. 223 (1984) 194–199. https://doi.org/10.1016/0167-5087(84)90646-X.

[25] J. Martínez, T. Baciu, A. Peñalver, C. Aguilar, F. Borrull, Occurrence of and radioanalytical methods used to determine medical radionuclides in environmental and biological samples. A review, J. Environ. Radioact. 207 (2019) 37–52. https://doi.org/10.1016/j.jenvrad.2019.05.015.

[26] D. Lariviere, V.F. Taylor, R.D. Evans, R.J. Cornett, Radionuclide determination in environmental samples by inductively coupled plasma mass spectrometry,

Spectrochim. Acta Part B At. Spectrosc. 61 (2006) 877–904. https://doi.org/10.1016/j.sab.2006.07.004.

[27] P. Lerch, M. Cosandey, Comparison of Various Procedures for the Simultaneous Measurement of 32P and 45Ca in Inorganic Salts by Liquid Scintillation Counting, in: S. Rothchild (Ed.), Adv. Tracer Methodol., Springer US, New York, NY, 1966: pp. 107–117. https://doi.org/10.1007/978-1-4684-8625-4_12.

[28] M. Betti, S. Giannarelli, T. Hiernaut, G. Rasmussen, L. Koch, Detection of trace radioisotopes in soil, sediment and vegetation by glow discharge mass spectrometry, Anal. Bioanal. Chem. 355 (1996) 642–646. https://doi.org/10.1007/s0021663550642.

[29] Y. Kobayashi, Elucidation of the Metabolic Pathways of Selenium and Arsenic by Analytical Toxicology, J. Health Sci. 56 (2010) 154–160. https://doi.org/10.1248/jhs.56.154.

[30] M. De La Guardia, Strategies for the rapid characterization of metals and organic pollutants in solid wastes and contaminated soils by using mass spectrometry, TrAC Trends Anal. Chem. 17 (1998) 263–272. https://doi.org/10.1016/S0165-9936(97)00104-0.

[31] H. Tuovinen, D. Vesterbacka, E. Pohjolainen, D. Read, D. Solatie, J. Lehto, A comparison of analytical methods for determining uranium and thorium in ores and mill tailings, J. Geochem. Explor. 148 (2015) 174–180. https://doi.org/10.1016/j.gexplo.2014.09.004.

[32] F.J. Krug, Métodos de Preparo de Amostras: Fundamentos sobre o Preparo de Amostras Orgânicas e inorgânicas para análise elementar, 1st ed., 2008.

[33] E.C. Vieira, M.Y. Kamogawa, S.G. Lemos, J.D.A. Nóbrega, A.R.D.A. Nogueira, Decomposição de amostras de solos assistida por radiação microondas: estratégia para evitar a formação de fluoretos insolúveis, Rev. Bras. Ciênc. Solo 29 (2005) 547–553. https://doi.org/10.1590/S0100-06832005000400007.

[34] Q. Jin, F. Liang, H. Zhang, L. Zhao, Y. Huan, Daqian Song, Application of microwave techniques in analytical chemistry, TrAC Trends Anal. Chem. 18 (1999) 479–484. https://doi.org/10.1016/S0165-9936(99)00110-7.

[35] K. Srogi, A Review: Application of Microwave Techniques for Environmental Analytical Chemistry, Anal. Lett. 39 (2006) 1261–1288. https://doi.org/10.1080/00032710600666289.

[36] J.M. Mermet, C. Trassy, A Plasma Torch Configuration for Inductively Coupled Plasma as a Source in Optical Emission Spectroscopy and Atomic Absorption Spectroscopy, Appl. Spectrosc. 31 (1977) 237–239. https://doi.org/10.1366/000370277774463751.

[37] J. Szpunar, Advances in analytical methodology for bioinorganic speciation analysis: metallomics, metalloproteomics and heteroatom-tagged proteomics and metabolomics, The Analyst 130 (2005) 442. https://doi.org/10.1039/b418265k.

[38] H.I. Mira, N.M. Alanwar Abdalaty, Disposal of Uranium and Thorium Entering the Human Digestive System before Reaching the Blood by Adsorption on Simethicone and Forming a Non-absorbable Compound: A Full in vitro Analysis, Indian J. Pharm. Educ. Res. 55 (2021) 146–152. https://doi.org/10.5530/ijper.55.1.1.

[39] S. Ito, Y. Takaku, M. Ikeda, Y. Kishimoto, Determination of trace levels of uranium and thorium in high purity gadolinium sulfate using the ICP-MS with solid-phase chromatographic extraction resin, Prog. Theor. Exp. Phys. 2017 (2017). https://doi.org/10.1093/ptep/ptx145.

[40] J. Kuwabara, S. Tolmachyov, H. Noguchi, The development of flow injection technique for rapid uranium determination in urine samples, J. Nucl. Sci. Technol. 39 (2002) 556–559. https://doi.org/10.1080/00223131.2002.10875529.

[41] P. Schramel, I. Wendler, P. Roth, E. Werner, Method for the determination of thorium and uranium in urine by ICP-MS, Mikrochim. Acta 126 (1997) 263–266. https://doi.org/10.1007/BF01242331.

[42] B.-R. Li, H. Tang, R.-Q. Yu, J.-H. Jiang, Single-Nanoparticle ICP-MS for Sensitive Detection of Uracil-DNA Glycosylase Activity, Anal. Chem. 93 (2021) 8381–8385. https://doi.org/10.1021/acs.analchem.1c01447.

[43] S.S. Poteshin, A.A. Sysoev, S.S. Lagunov, A. Sereda, V.V. Sosnovtsev, A.I. Bolozdynya, Y.B. Efremenko, Determination of Traces of Uranium and Thorium in Titanium and Copper Used for the Construction of the Russian Emission Detector 100 by Inductively Coupled Plasma Mass Spectrometry, Eur. J. Mass Spectrom. 21 (2015) 335–340. https://doi.org/10.1255/ejms.1314.
[44] P. Roth, V. H??llriegl, W.B. Li, U. Oeh, P. Schramel, VALIDATING AN IMPORTANT ASPECT OF THE NEW ICRP BIOKINETIC MODEL OF THORIUM:, Health Phys. 88 (2005) 223–228. https://doi.org/10.1097/01.HP.0000148546.80698.70.
[45] J. Avivar, L. Ferrer, M. Casas, V. Cerdà, Fully automated lab-on-valve-multisyringe flow injection analysis-ICP-MS system: an effective tool for fast, sensitive and selective determination of thorium and uranium at environmental levels exploiting solid phase extraction, J. Anal. At. Spectrom. 27 (2012) 327. https://doi.org/10.1039/c2ja10304d.
[46] P. Wang, E. Hu, Q. Wang, Z. Lei, H. Wang, Y. Zhang, W. Hou, R. Zhang, Selective extraction of uranium from uranium–beryllium ore by acid leaching, J. Radioanal. Nucl. Chem. 322 (2019) 597–604. https://doi.org/10.1007/s10967-019-06689-1.
[47] N. Veerasamy, S. Kasar, R. Murugan, K. Inoue, T. Natarajan, R. Chand Ramola, M. Fukushi, S. Kumar Sahoo, 234U/238U disequilibrium and 235U/238U ratios measured using MC-ICP-MS in natural high background radiation area soils to understand the fate of uranium, Chemosphere 323 (2023) 138217. https://doi.org/10.1016/j.chemosphere.2023.138217.
[48] D. Curtis, J. Fabryka-Martin, P. Dixon, J. Cramer, Nature's uncommon elements: plutonium and technetium, Geochim. Cosmochim. Acta 63 (1999) 275–285. https://doi.org/10.1016/S0016-7037(98)00282-8.
[49] J.A.M. Porto, L. De Jesus Alves, F.C. Nunes, E. Gross, H.H.S. Medrado, J.D.S. Rocha, M.M. Viana, A.P.D.C. Teixeira, M.N.V. Prasad, Nuclear energy and its role in decarbonization: Scenarios and perspectives, in: Decarbonization Strateg. Driv. Achieve Carbon Neutrality Sustain., Elsevier, 2024: pp. 97–120. https://doi.org/10.1016/B978-0-443-13607-8.00009-2.
[50] S. Grangeon, C. Roux, C. Lerouge, P. Chardon, R. Beuzeval, G. Montavon, F. Claret, T. Grangeon, Geochemical and mineralogical characterization of streams and wetlands downstream a former uranium mine (Rophin, France), Appl. Geochem. 150 (2023) 105586. https://doi.org/10.1016/j.apgeochem.2023.105586.
[51] S. Masod Abdulqader, B. Vakanjac, J. Kovačević, Z. Naunovic, N. Zdjelarević, Natural Radioactivity of Intrusive-Metamorphic and Sedimentary Rocks of the Balkan Mountain Range (Serbia, Stara Planina), Minerals 8 (2017) 6. https://doi.org/10.3390/min8010006.
[52] A.M.R. Neiva, P.C.S. Carvalho, I.M.H.R. Antunes, M.T.D. Albuquerque, A.C.S. Santos, P.P. Cunha, S.B.A. Henriques, Assessment of metal and metalloid contamination in the waters and stream sediments around the abandoned uranium mine area from Mortórios, central Portugal, J. Geochem. Explor. 202 (2019) 35–48. https://doi.org/10.1016/j.gexplo.2019.03.020.
[53] M. Fayek, J. Horita, E.M. Ripley, The oxygen isotopic composition of uranium minerals: A review, Ore Geol. Rev. 41 (2011) 1–21. https://doi.org/10.1016/j.oregeorev.2011.06.005.
[54] A.M. Spiridonov, L.D. Zorina, V.A. Romanov, Types of endogenous geochemical fields and their significance for prospecting, Russ. Geol. Geophys. 55 (2014) 290–297. https://doi.org/10.1016/j.rgg.2014.01.012.
[55] H.P. Schwarcz, L. Morawska, Uranium-Series Dating of Carbonates from Bir Tarfawi and Bir Sahara East, in: Egypt Last Interglacial, Springer US, Boston, MA, 1993: pp. 205–217. https://doi.org/10.1007/978-1-4615-2908-8_11.
[56] B. Li, W. Liu, Z. Wen, D. Su, Q. Shi, Study on Deep Uranium Information of Radon Anomaly Extraction Technology, Acta Geol. Sin. - Engl. Ed. 88 (2014) 1366–1367. https://doi.org/10.1111/1755-6724.12381_14.
[57] - Hmingchungnunga, - Vanramlawma, - Remlalsiama, - Laldingngheta, L.Z. Chhangte, Z. Pachuau, B. Zoliana, - Rosangliana, B.K. Sahoo, B.K. Sapra, Assessment of radon

content in water using SMART RnDuo in Mizoram, Northeast India, in: Proc. Mizoram Sci. Congr. 2018 MSC 2018 - Perspect. Trends Dev. Sci. Educ. Res., Atlantis Press, Aizawl, India, 2018. https://doi.org/10.2991/msc-18.2018.32.

[58] D. Francisca, W. Iramina, A. Ayres Da Silva, G. De Tomi, New Evaluation Method of Exposure to Radon Gas in Mining Environments, Minerals 13 (2023) 897. https://doi.org/10.3390/min13070897.

[59] H. Nisbet, A.A. Migdisov, A.E. Williams-Jones, H. Xu, V.J. Van Hinsberg, R. Roback, Challenging the thorium-immobility paradigm, Sci. Rep. 9 (2019) 17035. https://doi.org/10.1038/s41598-019-53571-x.

[60] Z. Li, M. Hadioui, K.J. Wilkinson, Conditions affecting the release of thorium and uranium from the tailings of a niobium mine, Environ. Pollut. 247 (2019) 206–215. https://doi.org/10.1016/j.envpol.2018.12.042.

[61] Y. Huang, Y. Liu, Y. Ruan, J. Gong, L. Kong, M. Su, W. Han, M. Yang, Husman·zaulem, D. Chen, A sulfonated ligand-aqueous two-phase system for selective extraction of thorium from mining wastewater: Process optimization, structural characterization and mechanism exploration, J. Mol. Liq. 384 (2023) 122223. https://doi.org/10.1016/j.molliq.2023.122223.

[62] C. Kamunda, M. Mathuthu, M. Madhuku, An Assessment of Radiological Hazards from Gold Mine Tailings in the Province of Gauteng in South Africa, Int. J. Environ. Res. Public. Health 13 (2016) 138. https://doi.org/10.3390/ijerph13010138.

[63] J.A. Ademola, Exposure to high background radiation level in the tin mining area of Jos Plateau, Nigeria, J. Radiol. Prot. 28 (2008) 93–99. https://doi.org/10.1088/0952-4746/28/1/006.

[64] W. M. Al-Areqi, SEPARATION AND RADIOLOGICAL IMPACT ASSESSMENT OF THORIUM IN MALAYSIAN MONAZITE PROCESSING, Malays. J. Anal. Sci. 20 (2016) 770–776. https://doi.org/10.17576/mjas-2016-2004-09.

[65] A. Cardenas, M.I. Dias, C. Diamantino, E. Carvalho, D. Russo, R. Marques, Geochemical Behavior of Lanthanides and Actinides in an Old Uranium Mine, Portugal, Geosciences 13 (2023) 168. https://doi.org/10.3390/geosciences13060168.

[66] N.D. Chau, P. Wyszomirski, E. Chruściel, A. Ochoński, Uranium in mining waters of kaolin open pit in arów (Lower Silesia); methodology of determination and genetic remarks, Appl. Radiat. Isot. 51 (1999) 593–597. https://doi.org/10.1016/S0969-8043(99)00072-X.

[67] F. Santos-Francés, E. Gil Pacheco, A. Martínez-Graña, P. Alonso Rojo, C. Ávila Zarza, A. García Sánchez, Concentration of uranium in the soils of the west of Spain, Environ. Pollut. 236 (2018) 1–11. https://doi.org/10.1016/j.envpol.2018.01.038.

[68] Y. Yuan, Q. Yu, J. Wen, C. Li, Z. Guo, X. Wang, N. Wang, Ultrafast and Highly Selective Uranium Extraction from Seawater by Hydrogel-like Spidroin-based Protein Fiber, Angew. Chem. Int. Ed. 58 (2019) 11785–11790. https://doi.org/10.1002/anie.201906191.

[69] S. Biswas, V.H. Rupawate, K.N. Hareendran, S.B. Roy, J.K. Chakravartty, Novel precipitation technique for uranium recovery from carbonate leach solutions, J. Radioanal. Nucl. Chem. 304 (2015) 1345–1351. https://doi.org/10.1007/s10967-014-3863-x.

[70] B.I. Omel'yanenko, V.A. Petrov, V.V. Poluektov, Behavior of uranium under conditions of interaction of rocks and ores with subsurface water, Geol. Ore Depos. 49 (2007) 378–391. https://doi.org/10.1134/S1075701507050042.

[71] F.P. Carvalho, J.M. Oliveira, M. Malta, Preliminary assessment of uranium mining legacy and environmental radioactivity levels in Sabugal region, Portugal, Int. J. Energy Environ. Eng. 7 (2016) 399–408. https://doi.org/10.1007/s40095-016-0219-z.

[72] M. Gavrilescu, L.V. Pavel, I. Cretescu, Characterization and remediation of soils contaminated with uranium, J. Hazard. Mater. 163 (2009) 475–510. https://doi.org/10.1016/j.jhazmat.2008.07.103.

[73] F.D. Tillman, K.R. Beisner, J.R. Anderson, J.A. Unema, An assessment of uranium in groundwater in the Grand Canyon region, Sci. Rep. 11 (2021) 22157. https://doi.org/10.1038/s41598-021-01621-8.
[74] V. Popovych, A. Voloshchyshyn, D. Rudenko, N. Popovych, Geochemical properties of water under the waste heaps in Chervonohrad mining region, E3S Web Conf. 123 (2019) 01035. https://doi.org/10.1051/e3sconf/201912301035.
[75] Y. Moliner-Martinez, P. Campíns-Falcó, P.J. Worsfold, M.J. Keith-Roach, The impact of a disused mine on uranium transport in the River Fal, South West England, J Env. Monit 6 (2004) 907–913. https://doi.org/10.1039/B409129A.
[76] M. Englert, L. Krall, R.C. Ewing, Is nuclear fission a sustainable source of energy?, MRS Bull. 37 (2012) 417–424. https://doi.org/10.1557/mrs.2012.6.
[77] D. Langmuir, J.S. Herman, The mobility of thorium in natural waters at low temperatures, Geochim. Cosmochim. Acta 44 (1980) 1753–1766. https://doi.org/10.1016/0016-7037(80)90226-4.
[78] M. Amrane, L. Oufni, Determination for levels of uranium and thorium in water along Oum Er-Rabia river using alpha track detectors, J. Radiat. Res. Appl. Sci. 10 (2017) 246–251. https://doi.org/10.1016/j.jrras.2017.05.002.
[79] A.-M. Hossu, M.G. Milotin, I. Ionita, A. Stoica, E. Barascu, The influence of the exploitation of uranium deposits on the soil and vegetation, in: M. Vladescu, I. Cristea, R.D. Tamas (Eds.), Adv. Top. Optoelectron. Microelectron. Nanotechnologies XI, SPIE, Constanta, Romania, 2023: p. 13. https://doi.org/10.1117/12.2642131.
[80] M. Fomina, J.W. Hong, G.M. Gadd, Effect of depleted uranium on a soil microcosm fungal community and influence of a plant-ectomycorrhizal association, Fungal Biol. 124 (2020) 289–296. https://doi.org/10.1016/j.funbio.2019.08.001.
[81] G. Echevarria, M.I. Sheppard, J. Morel, Effect of pH on the sorption of uranium in soils, J. Environ. Radioact. 53 (2001) 257–264. https://doi.org/10.1016/S0265-931X(00)00116-8.
[82] M. Stojanović, D. Stevanović, J. Milojković, M.L. Mihajlović, Z. Lopičić, T. Šoštarić, Influence of Soil Type and Physical–Chemical Properties on Uranium Sorption and Bioavailability, Water. Air. Soil Pollut. 223 (2012) 135–144. https://doi.org/10.1007/s11270-011-0845-2.
[83] M. Stojanović, L. Pezo, M. Mihajlović, J. Petrović, M. Petrović, T. Šoštarić, J. Milojković, Chemometric approach for prediction of uranium pathways in the soil, Radiochim. Acta 102 (2014) 701–710. https://doi.org/10.1515/ract-2013-2178.
[84] M. Antunes, A. Santos, T. Valente, T. Albuquerque, Spatial Mobility of U and Th in a U-enriched Area (Central Portugal), Appl. Sci. 10 (2020) 7866. https://doi.org/10.3390/app10217866.
[85] M. Stojanović, M. Mihajlović, Z. Lopičić, J. Milojković, T. Šoštarić, M. Petrović, The influence of soil type on maize and wheat uranium uptake, Qual. Assur. Saf. Crops Foods 5 (2013) 237–242. https://doi.org/10.3920/QAS2012.0229.
[86] F.Y.C. Huang, P.V. Brady, E.R. Lindgren, P. Guerra, Biodegradation of Uranium−Citrate Complexes: Implications for Extraction of Uranium from Soils, Environ. Sci. Technol. 32 (1998) 379–382. https://doi.org/10.1021/es970181d.
[87] C.F.V. Mason, W.R.J.R. Turney, B.M. Thomson, N. Lu, P.A. Longmire, C.J. Chisholm-Brause, Carbonate Leaching of Uranium from Contaminated Soils, Environ. Sci. Technol. 31 (1997) 2707–2711. https://doi.org/10.1021/es960843j.
[88] S.C. Träber, W.B. Li, V. Höllriegl, K. Nebelung, B. Michalke, W. Rühm, U. Oeh, Calculation of internal dose from ingested soil-derived uranium in humans: Application of a new method, Radiat. Environ. Biophys. 54 (2015) 265–272. https://doi.org/10.1007/s00411-015-0602-9.
[89] J. Nriagu, D.-H. Nam, T.A. Ayanwola, H. Dinh, E. Erdenechimeg, C. Ochir, T.-A. Bolormaa, High levels of uranium in groundwater of Ulaanbaatar, Mongolia, Sci. Total Environ. 414 (2012) 722–726. https://doi.org/10.1016/j.scitotenv.2011.11.037.
[90] V.P. Mironov, J.L. Matusevich, V.P. Kudrjashov, S.F. Boulyga, J.S. Becker, Determination of irradiated reactor uranium in soil samples in Belarus using 236U as

irradiated uranium tracer, J. Environ. Monit. 4 (2002) 997–1002. https://doi.org/10.1039/b207573c.

[91] H.M. Fernandes, F.F. Lamego Simoes Filho, V. Perez, M.R. Franklin, L.A. Gomiero, Radioecological characterization of a uranium mining site located in a semi-arid region in Brazil, J. Environ. Radioact. 88 (2006) 140–157. https://doi.org/10.1016/j.jenvrad.2006.01.006.

[92] M. Momčilović, J. Kovačević, M. Tanić, M. Đorđević, G. Bačić, S. Dragović, Distribution of natural radionuclides in surface soils in the vicinity of abandoned uranium mines in Serbia, Environ. Monit. Assess. 185 (2013) 1319–1329. https://doi.org/10.1007/s10661-012-2634-9.

[93] M.I. Abdul Adziz, K.S. Khoo, An assessment of absorbed dose and radiation hazard index from soil around repository facility at Bukit Kledang, Perak, Malaysia, IOP Conf. Ser. Mater. Sci. Eng. 298 (2018) 012001. https://doi.org/10.1088/1757-899X/298/1/012001.

Supplementary Information

Determination of Natural Uranium and Thorium Levels in Environmental Matrices Surrounding Uranium Mining Sites via ICP-MS

Joel Augusto Moura Porto[a], Lander de Jesus Alves[b], Hector Hugo Silva Medrado[c], Josilene da Silva Rocha[c], Rodrigo Gibaut de Souza Góis[c], Fábio Carvalho Nunes[d], Marcelo Machado Viana[a], and Ana Paula de Carvalho Teixeira[a]*

[a]Department of Chemistry, Federal University of Minas Gerais (UFMG), Belo Horizonte, Minas Gerais, Brazil

[b]State University of Santa Cruz (UESC), Postgraduate Program in Biology and Biotechnology of Microorganisms, Ilhéus, Bahia, Brazil

[c]Indústrias Nucleares do Brasil (INB) - Uranium Concentration Unit, Security, Radioprotection and Environment Management, Caetité, Bahia, Brazil

[d]Federal Institute of Education, Science and Technology of Bahia, Santa Inês, Bahia, Brazil

---

* anapaula.cta1@gmail.com

Table S1. Operating parameters adopted for PerkinElmer NexION® 300X ICP-MS.

Instrumental parameters	
Equipment used	PerkinElmer NexION® 300X ICP-MS
^{232}Th detection limit	< 0.5 ng.L^{-1} (ppt)
RF Power	1600W
Plasma gas flow	18 L min^{-1}
Auxiliary gas flow	1.2 L min^{-1}
Gas flow in the nebulizer	1.01 L min^{-1}
KED Mode	
input voltage	-3.00 V
Output voltage	-32.00 V
CRO[a]	-15.00 V
QRO[b]	-12.00 V
Data acquisition parameters	
Number of sweeps per read	50
Number of readings	1
Number of replicates	3
Dwell Time	25 s
Collision gas	-
RPq[c]	0.25 V
Gas flow in the Collision Cell	-
Calibration range	5.0 - 40.0µg L^{-1}

[a] CRO (Cell Rod Offset) – voltage applied to the quadrupole of the reaction/collision cell;

[b] QRO (Quadrupole Rod Offset) – voltage applied to the mass analyzer quadrupole;

[c] RPq relates the voltage applied to the quadrupole rods and needs to be optimized.

Figure S1. Location of collection points.

Figure S2. Location of collection points – Cachoeira Mine.

Figure S3. Location of collection points – Engenho Mine.

Figure S4. Calibration curve used in the determination of natural uranium.

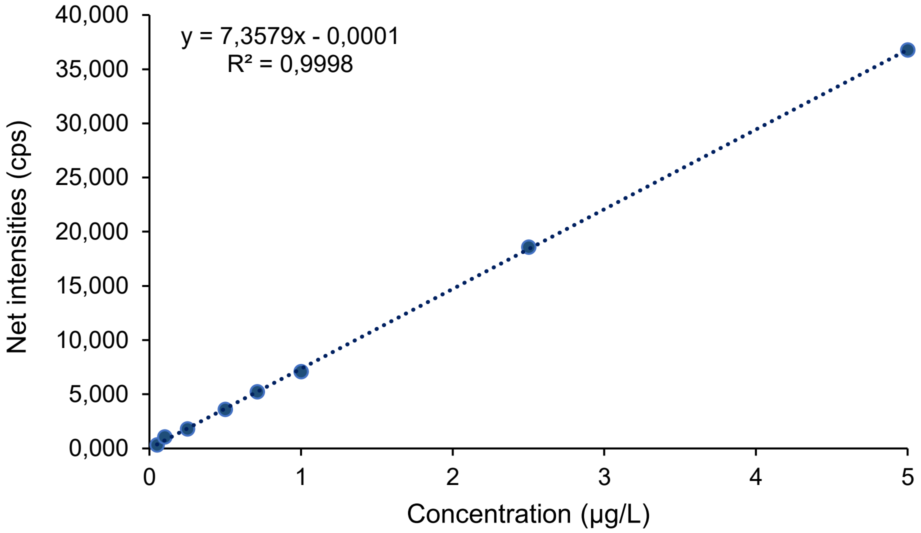
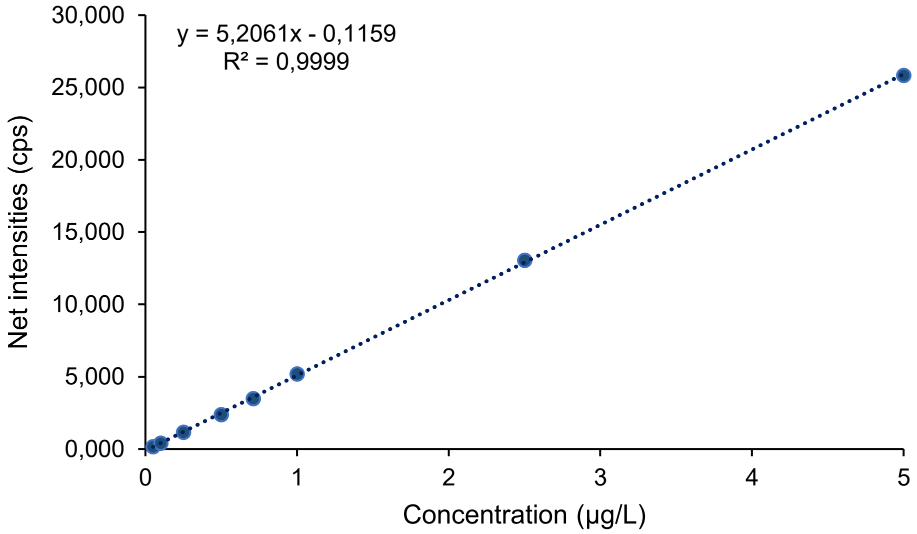


Figure S5. Calibration curve used in the determination of natural thorium.



Este preprint foi submetido sob as seguintes condições:

- Os autores declaram que os necessários Termos de Consentimento Livre e Esclarecido de participantes ou pacientes na pesquisa foram obtidos e estão descritos no manuscrito, quando aplicável.
- Os autores declaram que a elaboração do manuscrito seguiu as normas éticas de comunicação científica.
- Os autores declaram que estão cientes que são os únicos responsáveis pelo conteúdo do preprint e que o depósito no SciELO Preprints não significa nenhum compromisso de parte do SciELO, exceto sua preservação e disseminação.
- Os autores declaram que os dados, aplicativos e outros conteúdos subjacentes ao manuscrito estão referenciados.
- O manuscrito depositado está no formato PDF.
- Os autores declaram que a pesquisa que deu origem ao manuscrito seguiu as boas práticas éticas e que as necessárias aprovações de comitês de ética de pesquisa, quando aplicável, estão descritas no manuscrito.
- Os autores declaram que uma vez que um manuscrito é postado no servidor SciELO Preprints, o mesmo só poderá ser retirado mediante pedido à Secretaria Editorial do SciELO Preprints, que afixará um aviso de retratação no seu lugar.
- Os autores concordam que o manuscrito aprovado será disponibilizado sob licença Creative Commons CC-BY.
- O autor submissor declara que as contribuições de todos os autores e declaração de conflito de interesses estão incluídas de maneira explícita e em seções específicas do manuscrito.
- Os autores declaram que o manuscrito não foi depositado e/ou disponibilizado previamente em outro servidor de preprints ou publicado em um periódico.
- Caso o manuscrito esteja em processo de avaliação ou sendo preparado para publicação mas ainda não publicado por um periódico, os autores declaram que receberam autorização do periódico para realizar este depósito.
- O autor submissor declara que todos os autores do manuscrito concordam com a submissão ao SciELO Preprints.