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# Study of the relationship between mineralogical types and potential radon in rocks of the region of Serra do Gandarela – Quadrilátero Ferrífero-MG.

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## ABSTRACT

**The Quadrilátero Ferrífero (QF) is one of the most well-known metallogenetic provinces in the world and has its geological context well studied since the end of the 17th century. The Serra do Gandarela is positioned in the northeast of the QF and is supported by units of the Minas Supergroup. The Moeda Formation (MF) in the Serra do Gandarela hosts an uranium (U) occurrence which was recently characterized in terms of mineralogical context. Uraninite, coffinite and brannerite are the main U minerals present. It is expected that mineralogy and lithology contribute to radon emanation and exhalation, the radon (Rn) transport from the soil to the atmosphere. In this work, the Rn exhalation was correlated to the lithological types. Samples from the MF**

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**conglomerates and Nova Lima group schists were collected from the Serra do Gandarela. Rn exhalation measurements were performed on the samples with AlphaGUARD detector. These values were compared to the U content of samples obtained via INAA. The result of INAA was more expressive in the MF conglomerate sample from the NUCLEBRÁS drillcore, with a U concentration value of 24 µg.g$^{-1}$. The sample that had the highest average exhalation rate was also the same. The lowest value presented for both U concentration and average exhalation rate was shale, which were, 2 µg.g$^{-1}$ and 180 Bq.m$^{-3}$, respectively. The results are relevant for exploration and mining activities, during which Rn can accumulate in galleries. Additionally, the knowledge of the characteristic emanometric can be an important tool for geological and environmental studies.**

***Keywords:*** **radon exhalation, Moeda Formation, conglomerate, radiation detection, INAA.**

## 1. INTRODUCTION

The environment and the human being are constantly subject to a continuous flow of ionizing radiation from anthropogenic and natural sources. Primordial radionuclides have been present on Earth since its formation about 4.5 billion years ago. In this context, only radioactive isotopes whose half-life is long enough still exist naturally nowadays [1].

From the dosimetric and environmental point of view, the most important nuclides are potassium ($^{40}$K), uranium ($^{238}$U and $^{235}$U) and thorium ($^{232}$Th) decay series. The Th series and the two U series include radon isotopes, but the $^{238}$U decay series contains radon-222, the most important from a radiological or dosimetric viewpoint due to its abundance and relatively long half-life when it is compared with others radon isotopes. Epidemiological studies of human populations confirm the carcinogenic effect of radon and, nowadays, the International Agency for Research on Cancer classifies this radionuclide as a class I carcinogen [2]. For radon to be present in a given site, the radionuclides that originate it must be present in the soil and in the rocks that surround that region. In this way, soils rich in uranium and thorium are the main factor of the presence of this gas.

In the U decay series, radon is produced by the decay of radium (equation 1).

$$^{226}\mathrm{Ra} \rightarrow {}^{222}\mathrm{Rn} + \alpha \qquad (1)$$

The existence of radium in the mineral material leads to constant production of radon in every soil or aquifer. A fraction of the radon atoms is released from the solid matrix of the materials into

the interstitial space by a recoil as the radium decays [3]. This fraction is called Radon Emanation Coefficients (REC, known as coefficient of emanation, escape ratio, escape-to-production ratio, and percent emanation).

By diffusion and convection, such atoms are transported through the pores to the atmosphere where they are diluted [4]. A fraction of the radon atoms emanates from the site of their production into the porous space filled with soil, rock air (usually by diffusion or convection) or water, where some of them migrate and reach the surface and escape into the atmosphere (exhalation) [5].

The radon can be exhaled after emanation. The exhalation (E) studied in "closed exhalation circuit" can be found, in Becquerel, from the equation deduced as (equation 2):

$$E = \lambda C_{Rn} \frac{V}{S} \tag{2}$$

Where λ is the radon decay constant; $C_{Rn}$ is the radon concentration; V is the system volume and S is the sampling area. The exhalation and emanation of radon depend on factors such as porosity, grain size, distribution of radium in the atoms of the grain, among others [5].

The study of radon and its emanation rates from soils, rocks, and minerals has a wide range of applications in many branches of geosciences, e.g. in geological dating; as tracers to predict earthquakes [6]; to locate subsurface uranium ore and hydrocarbon deposits; and in atmospheric transport studies [7-8]. Then, is important to know these ratios to understand the physical and geological factors that affect radon release rates from rocks and minerals.

In this context, our purpose is to investigate the distribution of radioactivity in rocks in the Gandarela Syncline. We have conducted gamma scans in areas previously known from the literature for their uranium anomalies, and from samples of basal conglomerate of the Moeda Formation and schist of the Nova Lima Group, we measured the radon exhalation and uranium and thorium contents in those rocks.

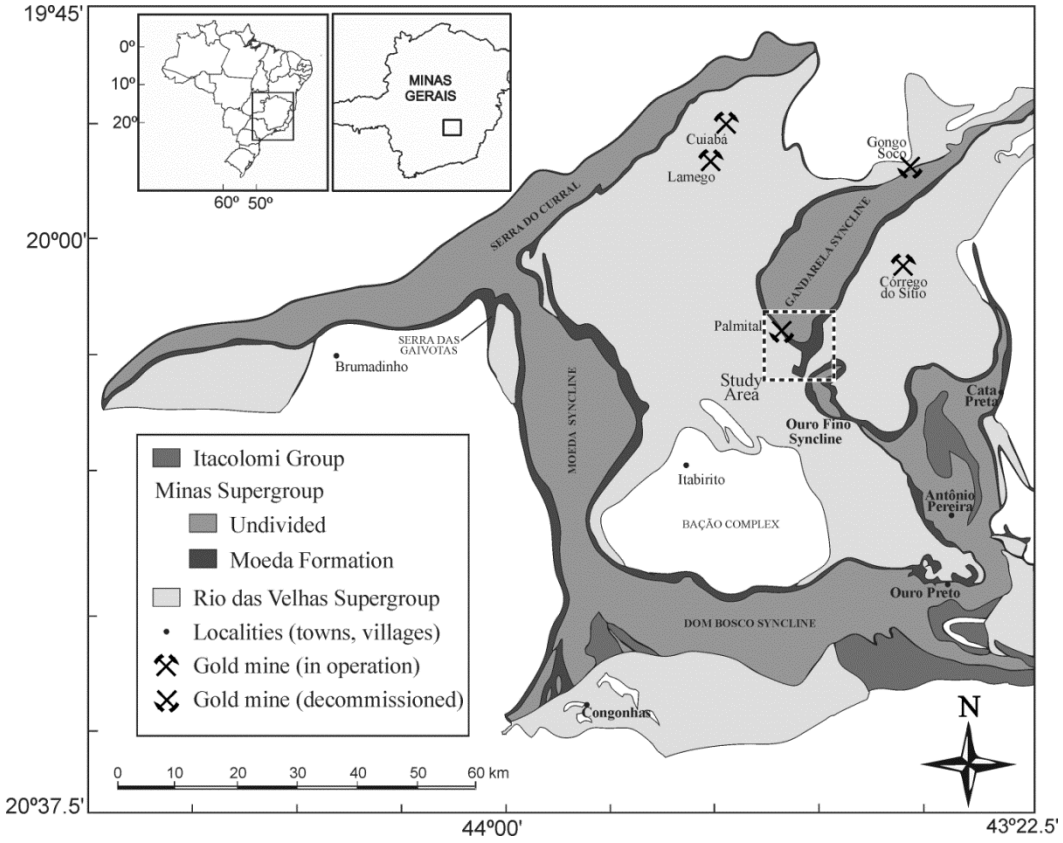
The information obtained can add general knowledge about the problems of indoor and outdoor radiation and their relationship with geology. The data can aid in the evaluation of future residential / commercial buildings in the area and in similar areas, as well as the risk of radon in mines that can be opened in the region and, finally, in the formulation of predictive models that identify potential areas with radon risk.

## 2. MATERIALS AND METHODS

### 2.1. Study area

The Quadrilátero Ferrífero (QF) comprises an area of approximately 15,000 km² (Figure 1). It is formed by ridges with altitudes from 1100 to 1600 meters [9]. These ridges are formed by folded metasedimentary sequences, especially the Minas Supergroup, which includes the largest iron deposits in the region [10]. It is located in the southern part of the São Francisco Craton [11].

**Figure 1:** *Simplified geological map of the Quadrilátero Ferrífero.*




Source: Based on Dorr (1969) and Pires (2012) in Guimarães (2019).

The Gandarela Syncline, Minas Gerais, Brazil, was the site selected for this study (Figure 1). Some radiometric anomalies were identified in this area in the 70's and 80's, and were discovered to be related to the basal metaconglomerates of the Moeda Formation, which overlays the shales of the Nova Lima Group [12]. The Moeda Formation has been extensively studied, but the

measurement of local radon and its correlation with the minerals in the region have not yet been widely studied.

The Moeda Formation in the Gandarela syncline is composed of polymictic and oligomictic metaconglomerates and coarse-, medium- and fine-grained quartzites. It overlaps the shales of the Nova Lima Group in discordant and erosive contact. The largest thickness occurs at the southern end of the syncline, reaching 250 meters, while the smallest occurs in the northern part where it is only 50 meters [13].

The Formation was divided into three stratigraphic units, each representing a sedimentation macrocycle [14]. Most of the uranium mineralization is associated to the matrix of the basal conglomerates, especially the ones with the highest maturity, oligomythic, pyritic and often carbonaceous, where uraninite and gold are present.

### 2.2. Experimental procediments

2.2.1 Fieldwork and gamma mapping

Fieldwork was carried out to perform the gamma mapping and to collect samples. Initially a radiometric survey with RS-230 BGO spectrometer (Figure 2) was performed. The RS-230 is manufactured in Canada and consists of an inorganic scintillator detector of Bismuth Germanate ($Bi_4Ge_3O_{12}$). The advantage of BGO in relation to Sodium Iodide (most commonly used for gamma radiation detections) is to have a high density and high atomic number which makes it very efficient for the detection of gamma rays. Measurements are performed in a span of 30 seconds.

**Figure 2:** *RS-230 BGO spectrometer*

Three sets of rock samples were collected : One set of basal Moeda Formation conglomerate (CNG01) and one set of Nova Lima shale (XNL01) by NUCLEBRÁS Gallery were collected from inside an old NUCLEBRAS gallery; One set of basal Moeda Formation conglomerate (CNG02) was obtained from a drillcore of NUCLEBRÁS uranium prospecting (Table 1).

**Table 1:** Sample's informations

| Sample set | Description | Origin | Justification |
|---|---|---|---|
| **CNG01** | Conglomerate (Moeda Formation). | NUCLEBRÁS Gallery. | Conglomerates from the Moeda Formation. |
| **CNG02** | Conglomerate (Moeda Formation). | Drillcore NUCLEBRÁS (20°7'51,9"S; 43°40'29,2"W) | The drillcore drilled by NUCLEBRÁS in the uranium prospecting campaign have the highest uranium mineralization. Drillcore information: 2SB69 box 58. |
| **XNL01** | Schist (Nova Lima Group). | NUCLEBRÁS Gallery. | The schist was collected from prospecting gallery (opened for the prospection of uranium). |

2.2.2 Petrography

Thin-sections of the conglomerate sample from the Moeda Formation were made in order to make a petrographical description of the rock. Petrography studies can show spatial distribution and minerals composition in samples placed on thin-section and allow for the evaluation of the presence of pyrite and organic matter in the conglomerates as described in other studies [15]. The petrographical studies were carried out using Leica petrographic optical microscopes, via transmitted and reflected light, with a coupled image capture system in the Mineralogical Characterization and Metalogenesis Laboratory CDTN / CNEN.

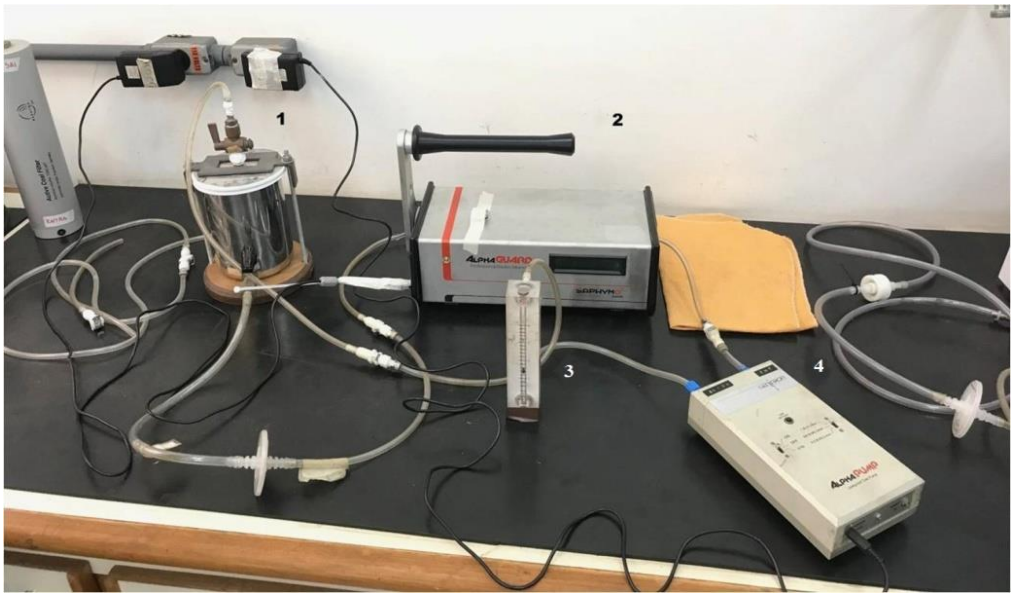
2.2.3 Methods for uranium, thorium and radon analysis

The uranium and thorium were quantified by Neutron Activation Analysis in the Neutron Activation Laboratory of CDTN / CNEN, using the TRIGA IPR-R1 reactor. Uranium was determined by Delayed Fission Neutron, a NAA method. The standard was $U_3O_8$ form IAEA, 0.471% and Th was determined by INAA, Relative method. The standard was $Th(NO_3)_4$ $5H_20$. This method is an adequate analytical method for the analysis of geological materials.

Radon exhalation was measured in the exhalation circuit built at the Natural Radioactivity Laboratory of the CDTN / CNEN. The detector used was the AlphaGUARD®, model PQ2000 PRO, manufactured by SAPHYMO GmbH, Germany (Figure 3) with an internal volume of 0.56 L and an internal potential of +750 V when connected. AlphaGUARD® uses an alpha spectrometer made out of an optimized pulse count ionization chamber where the gas diffuses into a glass fiber filter cylinder, which holds the radon decay products [16].

The samples are placed in the vessel (#1 in Figure 3), which was sealed with teflon to prevent the escape of radon gas. The pump (#4 in Figure 3) was operated in 1L/min to energize the circuit by circulating the gases. A rotor (#3 in Figure 3) was used to evaluate the flow of air flow.

**Figure 3:** *Exhalation circuit*



# 3. RESULTS AND DISCUSSION

## 3.1. Gamma mapping

The radiometric survey gamma counts varied from 40 to 2600 counts per second (cps). The cps levels were divided in very low (0-150 cps), medium (150-300 cps), high (300-450 cps), very high (450-2600 cps). Most of the surveyed area have low and medium levels of radiation. High and very high levels are associated with the occurrence of the Moeda Formation, particularly in the contat between Moeda Fm. and Nova Lima Group. Near this contact, the Moeda conglomerate has accumulations of various opaque minerals (mainly sulfides, of which pyrite predominates) with uranium and gold mineralizations [15]. The mean values of the radiometric data were plotted over the geological map of the Quadrilátero Ferrífero (Figure 4). The area delimited by the black square in Figure 4 was plotted in more detail in Figure 5.

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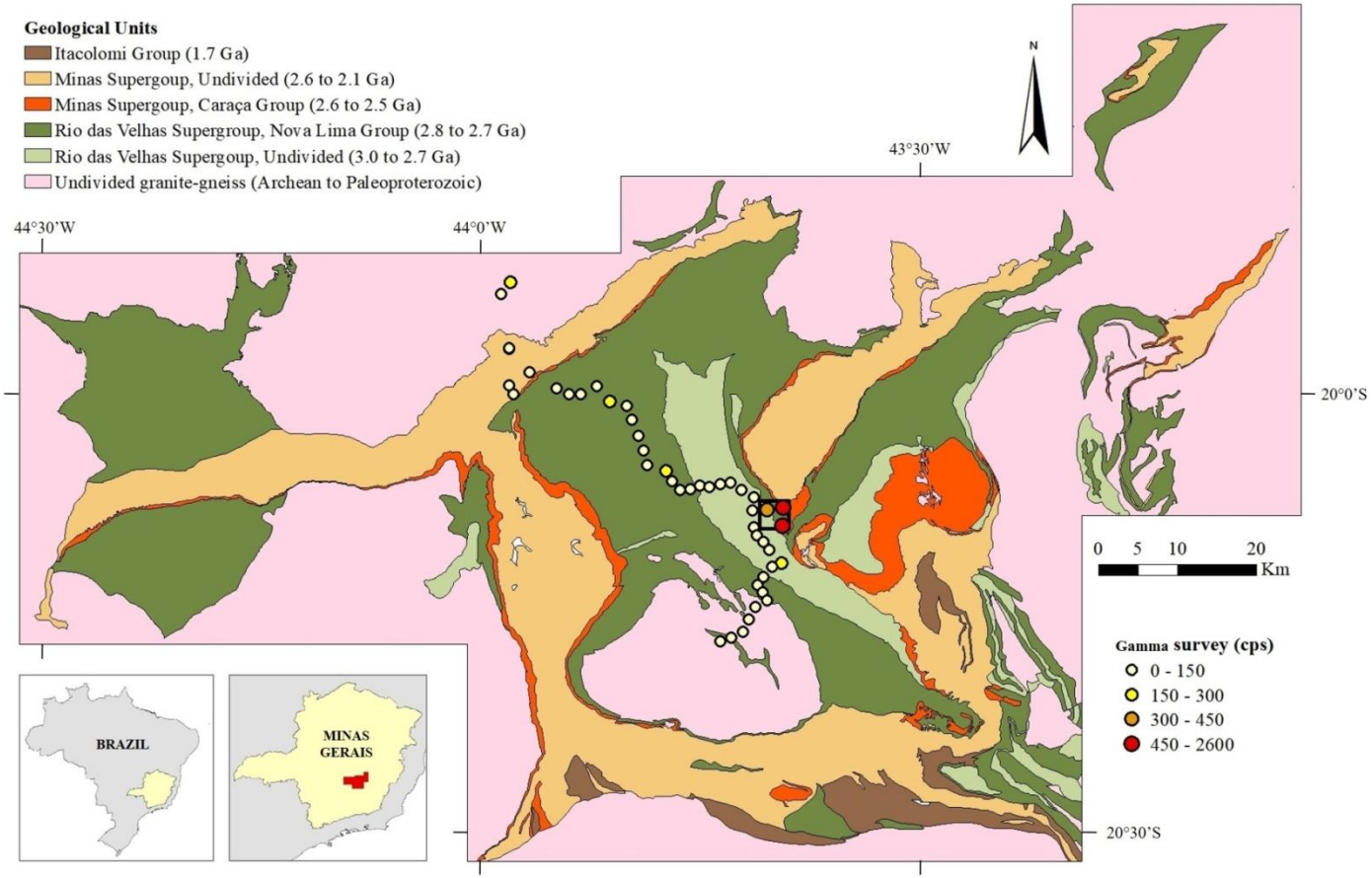
**Figure 4:** *Geological map of the Quadrilátero Ferrífero with gamma survey measurements*

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Source: *The geological map was modified from CODEMIG, based on Dorr II (1969).*

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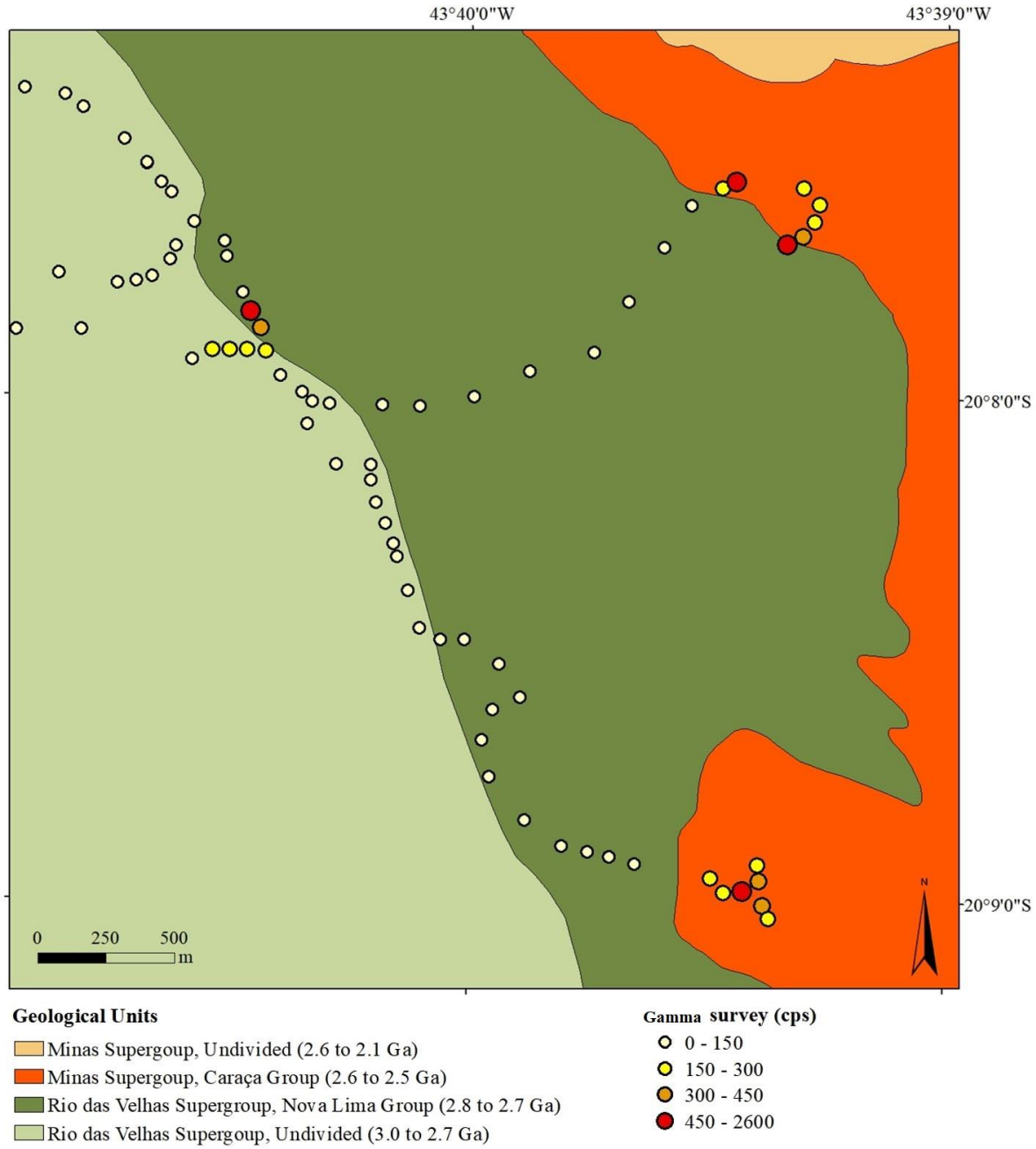
**Figure 5:** *Detailed map of the black square area in Figure 4*




Source: *Modified from CODEMIG based on Dorr II (1969).*

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### 3.2. Petrography

The petrographic study of the conglomerates that, geologically, are located at the base of the Moeda Formation, close to the erosive discordant contact with the Rio das Velhas Supergroup, is shown below (Figure 6). The rocks of the Rios das Velhas supergroup corresponding to the top of the Nova Lima Group occur as outcrops of gray quartz-muscovite shales, strongly deformed and with very fine granulometry.

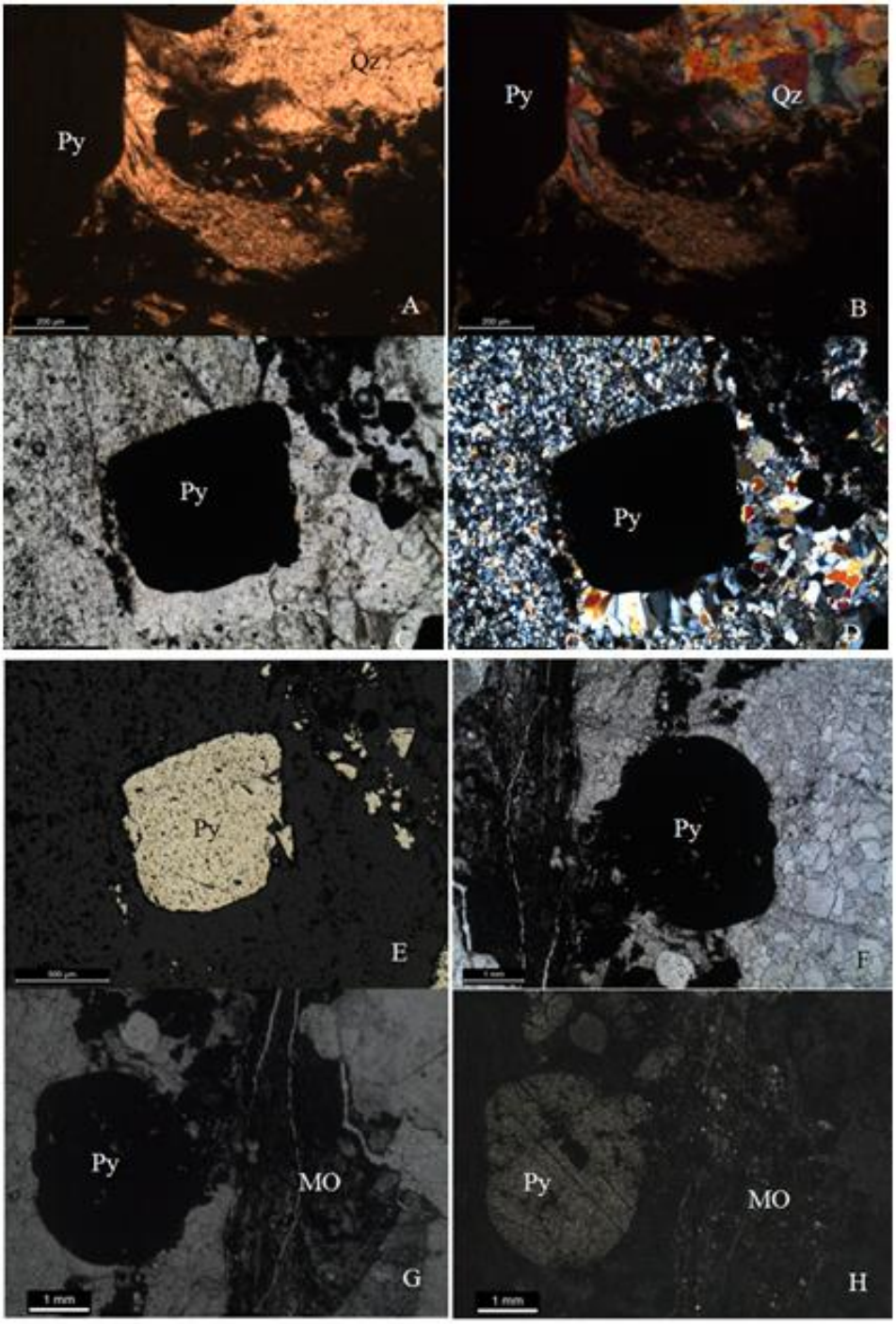
Petrography showed that the Moeda Formation conglomerate has characteristics similar to those described by Guimarães et al., (2019) [17], being constituted, in general terms, by an association of quartz matrix and organic matter (OM). The uraniferous ore occurs as micrometric brannerite ($UTi_2O_6$), uraninite ($UO_2$) and coffinite ($USiO_4$) either in the matrix or as inclusions in OM.

The matrix of the conglomerate of the Moeda Formation is predominantly quartz and presents micaceous domains of varied thicknesses. Pyrite ($FeS_2$) occurs in different generations and textures, ranging from round to euhedral. The crystals are fractured, sometimes porous, with mineral inclusions of quartz (Figure 6). Pyrites were classified into two types according to the classification proposed by Guimarães et al., (2019) [17]: (I) round and compact, represented by photomicrographs F, G and H (Figure 6); (II) porous pyrite using photomicrography C, D and E (Figure 6).

The organic matter is opaque and has a porous texture, (Figure 6 G, H) and its origins in the types of deposits are divergent. Some authors suggest that there is no direct correlation between OM and uranium mineralization [18]. More recent studies [19] show that organic matter functions as an important reducing trap for uranium present in oxidizing fluids. Barnicoat et al. (1997)[20] describes that the OM may have been formed after the formation of uraninite and brannerite, thus, the presence of the first mentioned would be due to the presence of radioactive minerals. However, Guimarães et al. (2019)[17] observed the occurrence of nodules of sterile organic matter in uraninite, brannerite or any other radioactive mineral.

Through this method it was observed semi quantitatively that the conglomerate has larger grains than those known in schist, as well as its porosity which is also greater in relation to schists.

**Figure 6:** *Petrography of the conglomerate Moeda Formation.*




A) and B) Photomicrography showing micaceous minerals; C), D) and E) Photomicrography shows quartz matrix (Qz) rich in pyrite (Py) and quartz fringes on its periphery. G) Photomicrography with transmitted light and H) Photomicrography with reflected light showing rounded pyrite and organic matter (MO). In the reflected light, it is possible to perceive details of the granulation and fractures of the pyrite.

### 3.3. Uranium, thorium and radon analysis

Uranium and thorium concentrations in the Moeda Formation conglomerate and Nova Lima Group shale obtained through neutron activation analysis are presented in the Table 2.

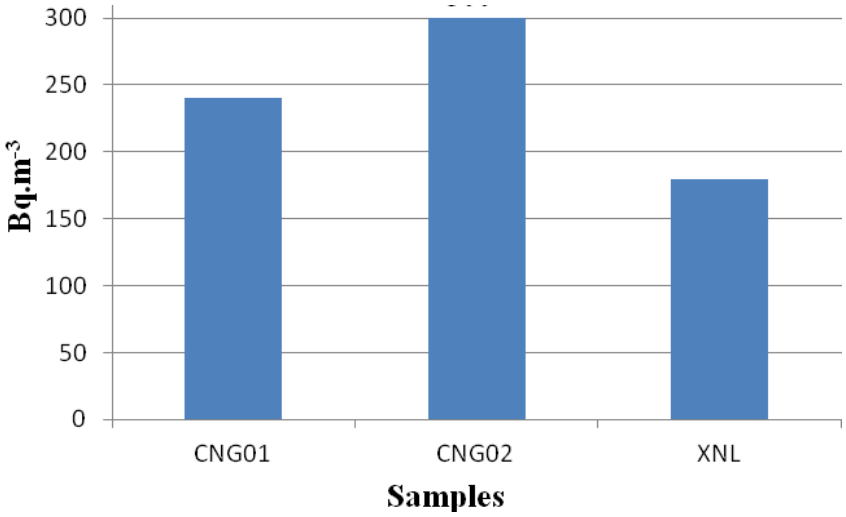
**Table 2:** U and Th results

| | **CNG01** | **CNG02** | **XNL** |
|---|---|---|---|
| **U (µg.g$^{-1}$)** | 16 ± 1 | 24 ± 4 | 2 ± 1 |
| **Th (µg.g$^{-1}$)** | 12.7 ± 0.6 | 42 ± 2 | 6.7 ± 0.3 |

The CNG02 has the highest uranium content. This result was expected, since the Moeda Formation conglomerate is known to be enriched in uranium and this sample presented the uranium mineralizations studied in the 1970's uranium prospecting. This can also be seen in the Th / U ratios: the CNG01 has a ratio of 0.79 and the has a ratio of CNG02 1.75 while the XNL has a ratio of 3.36. This ratio is very close to the Earth's crust average which is between 3.5 and 4 [21].

The radon gas exhalation was obtained by monitoring the temporal evolution of the gas in a closed circuit. In Figure 7 it is possible to observe the compilation of measurements in a column graph to each sample.

**Figure 7:** *Average radon concentration in the circuit*



In general, the studied rocks showed low radon exhalation values. Azevedo et al. (2015)[22] and Amaral et al. (2011)[23] working with ornamental rocks, in Ceará and Espírito Santo, respectively, concluded that the radon generated in the rock cannot reach the surface, since a large part of the gas is retained in the crystalline composition of the minerals in the rock**.** The table 3 shows the compilation of exhalation rate and compared with the concentrations obtained in neutron activation.

**Table 3:** Comparison of results

| **Sample** | **Rn Exhalation ratio ($\mu Bq.\ m^{-2}.s^{-1}$)** | **U content ($\mu g.\ g^{-1}$)** | **Ratio of radon exhalation by uranium content ($Bq.\ m^{-2}.s^{-1}$)/ ($\mu g.\ g^{-1}$)** |
|---|---|---|---|
| CNG01 | 15,4 | 16 ± 1 | $0{,}96E^{-6}$ |
| CNG02 | 27,6 | 24 ± 4 | $1{,}1E^{-6}$ |
| XNL | 10,9 | 2 ± 1 | $5{,}5E^{-6}$ |

The exhalation rate of radon was higher in the conglomerate samples than in the shale sample. Radon gas exhalation is not always related to the amount of uranium in the rock [22]. Possible explanations for this are the differences in porosity and grain size between shale and conglomerate, factors that are known to interfere with emanation and exhalation [22-23].

## 4. **CONCLUSION**

Gamma mapping showed 40 to 2600 cps throughout the area. The largest count is related to the gallery, where there are levels of the basal conglomerate of the Moeda Formation which is mineralized with uraninite, brannerite and coffinite in contact with the Nova Lima Group schist. The fundamental purpose of radon mapping is to prevent the population from being exposed to high values of gas concentration.

The Moeda Formation conglomerates, CNG01 and CNG02, had an average Rn concentration of 240 $Bq.m^{-3}$ and 300 $Bq.m^{-3}$ respectively and the Nova Lima (XNL) shale of 180 $Bq.m^{-3}$. The determination of radon exhalation rates allowed us to verify that the presence of uranium minerals does not always determine, or is proportional to, the amount of radon emanated and consequently

exhaled. For this purpose it is necessary to consider petrographic and textural factors of the rocks and their positioning in the field.

The petrography of the conglomerate is in agreement with data obtained from equivalent samples already published in previous geological studies: they present a quartz matrix, with the presence of organic matter and a wide variety of iron sulfides (pyrites) from different generations.

Countries should implement national mapping programs to determine areas subject to a greater risk of exposure to radon, preferably through studies with geological and geographic bases. It is possible to conclude that although the Gandarela Syncline is the main uraniferous area of the Quadrilátero Ferrífero (along with Serra das Gaivotas), it is radon risk can be considered low to medium and the results of this work can be used to compose studies on natural radioactivity in Serra do Gandarela- Quadrilátero Ferrífero-MG.

Therefore, our future studies are focused on understanding more factors and processes that cause changes in radon emanation and Study the exhalation of rocks indicating greater radioactivity in Belo Horizonte and the Metropolitan Region, conducting a detailed study of the contribution in these doses to indoor environments.

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