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ASSESSMENT OF RADON AND ITS PROGENY CONCENTRATION IN BRAZILIAN UNDERGROUND MINES

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ABSTRACT

Rock, soil and water contain ^{238}U, ^{232}Th and their decay products. The distribution of these radionuclides differs in terms of activity concentration depending on the mineral type and origin. All ore processing releases long and short half-life radionuclides, mainly radon and its progeny. It is important to monitor this gas and its products in underground mines in order to assess the radiological hazards of the exposed workers. On this concern, the present work outlines the characterization of the radon and its progeny concentration in the brazilian underground mines. The radon concentration was measured by using E-PERM Electrets Ion Chamber, AlphaGUARD and CR-39 track etch detectors. The radon progeny was determined by using DOSEman detector. The equilibrium state between radon and its progeny was calculated. Based on these data, the annual effective dose for miners was estimated according to UNSCEAR 2000 methodology. Moreover, the contribution from the main sources to the radon level inside mines was evaluated. For this, the following detectors were used: concentration measurements of the soil gas radon were carried out by AlphaGUARD detector; ^{226}Ra (^{214}Bi) specific activity in ore and soil samples were determined by gamma-ray spectrometry HPGe detector; and radon concentration in groundwater samples was performed by RAD7 detector. The radon concentration ranged from 113 to 8171 $Bq.m^{-3}$ and the Equilibrium Equivalent Concentration varied from 76 to 1174 $Bq.m^{-3}$. The general measurement results and the estimated effective doses will be used as bases to prepare the Brazilian regulatory standards.

1. INTRODUCTION

Uranium and thorium are present in the main rock minerals, incorporated in the secondary and accessory minerals and found in high concentration forming their own minerals [1]. These radioelements' distribution differs in terms of activity concentration depending on the mineral type and origin [2]. High background areas are found around the world and are the result of local geological controls and geochemical effects [3].

Thus, all mineral processing releases long and short half-life radionuclides from uranium and thorium series [4]. Miners face internal exposure to radon, thoron, and their short-lived decay products through inhalation as well as external exposure to gamma and beta radiation. However, the short-lived radon decay products are recognized as the main radiation health risk in mines [5].

The radon is produced by radium decay in ore bodies and by recoil emanating from the solid matrix of the material into the air- or water-filled pore space, or to rocks fractures [6]. When underground mines are opened, the radon is transported, by diffusion and convection, from the rocks to the galleries through water or air circulation. The importance of each process depends on the geological and tectonic fractures of the formation and of the hydrologic behavior of the aquifer [5]. Radon concentrations vary substantially in the atmosphere of underground mines depending on: the type of mine, the geological formation, the porosity, the moisture, the radon exhalation rate which varies due to alterations in the differential air pressure (naturally or mechanically), the uranium and radium levels, the working conditions, and the degree of ventilation [2, 5, 7,8].

The underground mine environment is complex and variable. Radon, after being exhaled, migrates along ventilation currents while it generates the solid decay products: ^{218}Po, ^{214}Pb, ^{214}Bi e ^{214}Po [5]. These radionuclides attach to the aerosol particles in the air, forming what is termed attached radon progeny. The fraction of radon progeny that does not attach to the aerosol particle in the air is termed the unattached state. In both cases, concentrations of these radon decay products increase rapidly with residence time of the air in the mine. If inhaled, both attached and unattached radon progenies may be deposited in the lung, especially in the upper respiratory tract, and irradiate the lung tissue as they decay. The entry of radioactive aerosol into the respiratory tract depends on their size; larger particles stop in the nasal cavity, while smaller aerosols reach the lungs [9]. Epidemiological studies have revealed a strong correlation between lung cancer and exposure to radon. It was identified as a human lung carcinogen in 1986 by the World Health Organization – WHO [10].

In this way, many surveys involving radon concentration measurements in underground mines have been conducted [2,4,6,7,8,11,12,13,14]. It is important to monitor this gas and its products in underground mines in order to assess the radiological hazards of the exposed workers. The knowledge of the radon distribution and its origin in mines is essential according to radiation-protection standards.

To address this concern, the authors aim to characterize the radon and its progeny concentration in 6 Brazilian underground mines. This work is a part of the Radon Project which is being conducted by the Nuclear Energy National Commission - CNEN. In Brazil, the National Department of Mineral Production reports the existence of the 72 underground mines in the country. A lot of these mines have uranium and thorium associations [11]. The

set of data will contribute to limit occupational radiation exposure of workers and to obtain information and parameters to prepare the Brazilian regulatory standards.

2. MATERIALS AND METHODS

2.1. Study Mines

The radon concentration was determined in 6 Brazilian mines. These mines include agalmatolite, coal, emerald, tourmaline, scheelite and fluorite extraction. The points selected for sampling consist of the active areas of each mine, in which we can observe the real working conditions of the miners at the date of collection. Drilling, detonation, transportation of materials, and maintenance of equipments and of the mine structure are examples of the typical activities observed in these mines [2].

2.2. Air Radon Measurement

The radon concentration measurements were performed by two detectors: E - PERM® electrets ion chamber (Radelec, Inc) and CR-39 nuclear track detector (Landauer).

E –PERM Electret Ion Chamber (Radelec, Inc.) consists of a stable electret (a charged Teflon disk) and a plastic chamber, acting as a passive integrating ionization monitor. In this detector, the radon passes through an input filter by diffusion until its concentration inside the chamber is equal to the analyzed environment. The radiation emitted by radon and its progeny ionizes the chamber air volume; the negative ions are attracted by positive electret surface, decreasing the initial voltage and the positive ions are dissipated by the wall chamber. Therefore, the radon concentration was estimated from the reduction in the electret potential surface, which is proportional to radon concentration integrated in exposure time. The detector's reading was performed by using the SPER-1 electronic device (Radelec, Inc). The voltage difference obtained was converted into radon concentration by using WINSPER Data Base and a Pre-programmed Palm Pilot software [15]. These detectors were placed in 2 - 7 points in each mine in order to perform initial measurement. Such measurement makes it possible to know the radon concentration within a mine. It is simple, produces quick results, and allows for decision-making. Radon measurements were taken at each point for at least two days (short-term measurement), according to a short-term measurement protocol of the Environmental Protection Agency [16,17].

E-PERMs are also sensitive to ions produced by radiation γ or X [17]. Then, the gamma exposure was determined by using Identifinder (THERMO) monitor in all sampling points selected. The readings are given in $\mu Sv.h^{-1}$. The mean value was first converted to $\mu R/h$ and then to a radon equivalent by E-PERM EIC specific constants. The result obtained was subtracted from the radon concentration acquired with electrets [15].

The CR-39 nuclear track detector is made up of allyl diglycol carbonate plastic [7]. The detectors (size 1x1.5cm) were attached to the cover of the diffusion chamber, a cylinder with a height of 2 cm and a diameter of 4 cm. In this system, the radon passes through an input filter which prevents dust particles from entering. The alpha particles, emitted from the radon

and its progeny, leave tracks in the film due to the Coulomb interactions with the materials atoms. In each mine, 2-8 points were selected along their full lengths, from the entrance of fresh air to its exhaust point. The detectors were exposed for approximately 3 months, according to a long-term measurement protocol of the Environmental Protection Agency [16,17]. After this time, the detectors were collected, covered in aluminum foil, and returned to the Natural Radioactivity Laboratory – LRN/CDTN. The CR-39 detectors were removed from the chamber and chemically etched with a 6.25 M solution of NaOH+ 2% of alcohol at 75^{o} C for 14 hours. The background was determined by processing an unexposed detector under identical conditions. The detectors were scanned using an optical microscope (ORTHOLUX) at 5x objective lens coupled to a DFC295 camera (Leica Microssystems Ltd.). For each detector, 15 images were made using LASV3.8 software. The set of images was processed using QUANTIKOV software, which provides the track density per cm^2. A conversion factor obtained in calibrated systems allows the conversion of the track density to radon concentration [18]. In this present work, the calibration factor (49 tracks $cm^{-2}kBq^{-1}m^3d^{-1}$) was determined by exposing the chamber arrangement in different radon concentration into calibration systems of the LRN/CDTN that are equipped with ^{226}Ra sources (NIST).

2.3. Equilibrium Factor and Effective Dose

In the dose evaluation, in general, the radon concentration is measured and an equilibrium factor (F) between radon and its progeny is assumed to be 0.4 [19, 20, 21]. However, the concentration of radon and its progeny may vary considerably within the mine with time and place [5,19]. Therefore, the adoption of a mean F is not recommended [2,19].

Included in this global concern, the radon concentration measurement was carried out with Alpha*GUARD*: a Saphymo GmbH system, model PQ2000PRO, that operates like a pulse ionization chamber. The monitor was programmed in diffusion mode at intervals of 10 and 60 minutes, acting as a continuous passive detector. Records of date, time, temperature, relative humidity and barometric pressure were also taken. The Alpha*GUARD* detection limit is 2 Bq/m^3 [2]. The data were acquired and treated with DataExpert software.

The short-lived radon progeny concentration is given in terms of the Potential Alpha Energy Concentration (PAEC). The PAEC can be expressed as Equilibrium Equivalent Concentration (EEC) [21]. PAEC and EEC measurements were carried out by alpha spectrometry using a solid state alpha detector, DOSEman PRO (Sarad). The detector was programmed at intervals of 60 minutes. The data were acquired and treated with Doseman 1.16 software.

The Alpha*GUARD* and DOSEman were installed together in underground mines for approximately 2 days. The sampling points were places where miners had maximum occupancy time and the equipments remained safe. Temperature (T) and relative humidity (RH) were also measured at these points.

The Equilibrium Factor - F (ratio of the EEC to the activity concentration of the radon) was estimated using the following formula [21]:

$$F = \frac{C_e}{C_o}, \quad (1)$$

where, *Ce* is the Equilibrium Equivalent Concentration and *Co* is the radon activity concentration in the air.

The effective dose for radon and its progeny was calculated using methodology from the UNSCEAR 2000 Report, which is derived from epidemiological studies and physical dosimetry [20]:

$$H = C_o \times F \times T \times k, \quad (2)$$

where, *T* (hours per year) is the occupancy in mines, *T*=2000 h [21] and *k* is the conversion factor, *k* = 9 nSv $(Bq.h.m^3)^{-1}$ established value used by UNSCEAR effective dose calculation [20].

2.4. Radon Measurement in Soil Gas

In order to measure the radon concentration in soil gas, sampling points were defined on the surface, directly on top of the mine galleries. The soil radionuclide concentration depends on the radioactive content of the rocks that originate them (geological process) and on geochemical causes [3]. Therefore, it is important to monitor radon in soil gas in order to assess the contribution from main radon sources in mines.

Soil gas radon concentration measurements were made by Alpha*GUARD* PQ2000PRO, a pulse ionization chamber, in flow mode at intervals of 1 minute. In this case, the technique consisted of positioning a metal probe into the soil to a depth of 70 cm in open circuit.

2.5. Measurement of ^{226}Ra Activity Concentrations

Samples of the different geological material that make up each mine were collected. This analysis aims to obtain information to support the study of the origin of radon levels inside mines.

The rock and soil samples were powdered, sieved and transferred to Marinelli beakers (1L). Each recipient remained tight sealed for 30 days in order to restore secular equilibrium between ^{226}Ra and its progeny.

The analytical technique used for ^{226}Ra (^{238}U chain) was gamma spectrometry, using a hyper-pure germanium detector (HPGe), coaxial geometry and 15% efficiency - a system from *CANBERRA*. The detector is located in a special laboratory with one of the lowest backgrounds in the world. The data were acquired and treated with *Genie 2000* software. Considering the equilibrium, ^{226}Ra activity concentration was measured by gamma-ray lines of ^{214}Bi (609,3 keV – 46,3%). In addition, the equilibrium condition was observed by measuring the gamma-ray lines of some natural series members, for example, ^{214}Bi and ^{214}Pb for ^{238}U.

2.6. Groundwater Radon Measurement

Groundwater radon concentration measurements were made by RAD7 (Durridge Company Inc.) to obtain the radon origin inside mines.

The RAD7 is a solid state silicon alpha detector. It requires an accessory kit, denominated RAD H_2O, in closed circuit, to measure water radon concentration. Wat-40 and Wat-250 protocols make calculations in 40 mL and 250 mL, respectively. We used the Wat-250 protocol that has one cycle constituted of five recycles: the first step is to aerate the water under controlled conditions and the others 4 intervals are to measure polonium activity (alpha spectrometry). The radon concentration is calculated based on its progeny.

The groundwater should be collected in special conditions: directly from the rocks, without contact with air.

3. RESULTS AND DISCUSSION

The radon concentrations (average, maximum and minimum) obtained with the EPERM and CR-39 detectors for each mine are presented in Table 1. It was found that radon concentration varies considerably from mine to mine and within the same mine. Many surveys involving radon concentration measurements in underground mines have verified this phenomenon [2,7,11]. Such results are a consequence of some factors being different in each mine including: specific activities of the radon's precursors in rocks and soil; permeability and porosity; ventilation and parameters such as temperature, humidity and pressure. According to Fathabadi *et al* (2005), the most important factor is ventilation (natural or mechanical), because it is different in all mines and at each point within a mine. As shown in Table 1, all mines except for mines B and F have points where radon concentration exceeded the proposed action level by the International Commission for Radiological Protection (ICRP) of 1000 $Bq.m^{-3}$ for a working place. This must be taken into account in monitoring and control [5]. It was noted that higher concentrations were verified in the middle and end galleries because of the transport of radon enriched air from the entrance of the mine to the air exhaust.

The radon concentration measured with AlphaGUARD, the radon progeny concentration measured with Doseman, and the equilibrium factor determined for each mine, are shown in Table 2, highlighting the mean value of the magnitudes of each mine. As can be observed, the equilibrium ranged from 0.2 to 0.7, with a mean value of 0.4. This wide range of values indicate that adopting the average for equilibrium factor in order to assess a dose is not

recommended. It was also found that mine A had different equilibrium factors when out of operating and in operation, which are 0.3 and 0.5, respectively. There is an explanation: certain mining activities such as detonation generates large amounts of aerosols inside galleries. Even though mine B has lower radon concentration, it has the highest equilibrium factor. Temperature and relative humidity range from 19-27^{o}C and 60 - 99%, respectively.

The effective dose to miners was calculated using Equation 2 and the values range from 1-21 mSv.a^{-1} (Table 2). The annual effective dose is a limited to range of about 3-10 mSv.a^{-1}[21]. As shown in Table 2, in two mines (mine A out of operating and mine E) the effective dose estimates are above the highest level suggested by ICRP. In addition, the effective dose to miners in these mines exceeds the limit of 20 mSv.a^{-1} for occupationally exposed individuals in Brazil [22].

Table 1: Radon Concentration obtained by EPERM Short-Term Measurement and CR-39 Long-Term Measurement in Underground Mines in Brazil

Mine	Mine Situation	Radon Concentration (Bq.m^{-3})	
		E-PERM [mean (max-min)]	CR-39 [(max-min)]
Mine A	Out of operaring	1339 (522-2616)	762 (599-1144)
Mine A	Operating	710 (692-728)	778 (507-1160)
Mine B	Operating	296 (180-625)	122
Mine C	Operating	2132 (689-3121)	985 (341-2913)
Mine D	Out of operaring	1311	787 (387-1256)
Mine E	Operating	5201 (2845-8171)	4153 (763-7384)
Mine F	Operating	292 (177-505)	316 (285-339)

Fig. 1 shows one example of the continuous monitoring of the radon and its progeny concentration in Mine A in operation performed at a selected point inside the mine in order to determine the equilibrium factor. There were some moments of high radon concentration, reaching up to 2088 Bq.m^{-3}. This may be explained by detonation activity that happened at approximately 5 pm and by the lack of ventilation overnight.

Table 2: Radon Concentration (AlphaGUARD), Equilibrium Equivalent Concentration (Doseman), Equilibrium Factor and Effective Dose in Brazilian Underground Mines

Mine	Mine Situation	Radon Concentration ($Bq.m^{-3}$)	Equilibrium Equivalent Concentration ($Bq.m^{-3}$)	Equilibrium Factor F	Effetive Dose ($mSv.a^{-1}$)
Mine A	Out of operating	3889	1174	0,3	21
Mine A	Operating	714	377	0,5	7
Mine B	Operating	113	76	0,7	1
Mine C	Operating	949	259	0,3	5
Mine D	Out of operating	1442	228	0,2	4
Mine E	Operating	4964	1148	0,2	21
Mina F	Operating	327	141	0,4	3

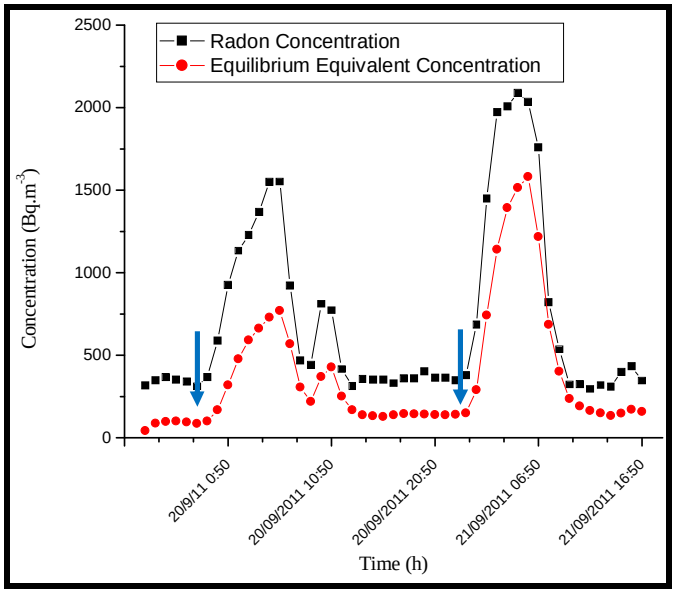

Figure 1: Example of the Continuous Monitoring of the Radon and its progeny Concentrations in the Mine A in operation. The arrows demonstrate the effect of detonation for radon activity within mine.

These considerations show points with enhanced radon concentration. In order to verify this enhanced concentration, the results of the ^{214}Bi (^{226}Ra) rock and soil specific activity, of the soil gas radon concentration and of the groundwater radon concentration are presented next.

The measured specific activities of ^{226}Ra in rocks and soils samples collected in the Mines A, B, C, D, E and F are presented in Fig. 2, Fig. 3, Fig. 4, Fig. 5, Fig. 6 and Fig. 7, respectively. The comparison of these values with the worldwide average concentration of ^{226}Ra in soil, reported by UNSCEAR (2000) as 32 Bq.kg^{-1} is shown in each Figure. The Fig. 8 shows the groundwater radon concentration of the surveyed mines. Figure 9 shows the soil radon concentration of each mine next to the typical range of 4,0 to 40,0 kBq.m^{-3} [23] of radon which occurs naturally in soils.

In mine A and C all samples collected had a significant ^{226}Ra specific activity (Fig. 2 and Fig. 4). It was also possible to verify that this mine has higher radon concentration in the groundwater (Fig. 8). This justifies the points with enhanced radon concentration in such mines.

In mine B, D and E only the pegmatite samples of different origin presented ^{226}Ra activity above the world average value in soil (Fig.3, Fig. 5 and Fig 6). In mine F no sample had enhanced ^{226}Ra activity (Fig. 7). They also did not have high groundwater radon concentration (Fig. 8). Therefore, the points with enhanced radon concentration in mines D and E may be a consequence of the pegmatite radon exhalations. Mines B and F presented relatively lower radon levels when compared to other values obtained in studied mines. This can be explained because mines B and F are smaller than the others. They have shorter ventilation pathways and, consequently, better ventilation and lower radon concentration. Even though mine B has pegmatite samples with significant ^{226}Ra activity.

Fig. 9 demonstrates that only mines A and B have soil radon concentrations above the typical range of 4,0 to 40,0 kBq.m^{-3}. This can be a result of the surrounding bedrock which produces the soil of the mines' regions.

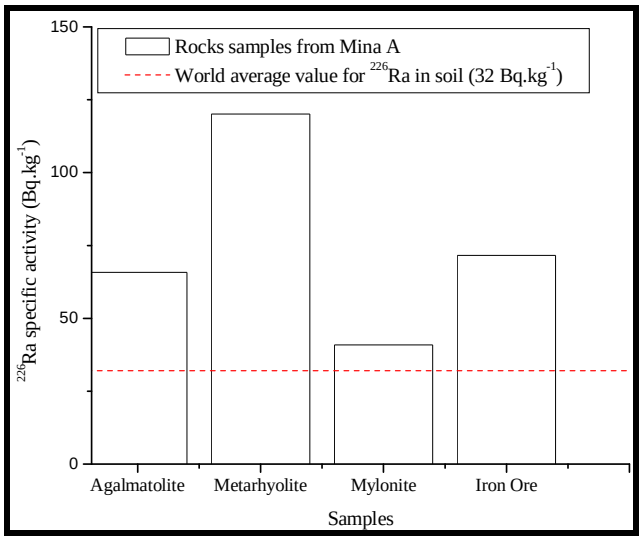

Figure 2: ^{226}Ra specific activities in rock samples from mine A. The comparison between the specific activities obtained and the ^{226}Ra worldwide average in soil.

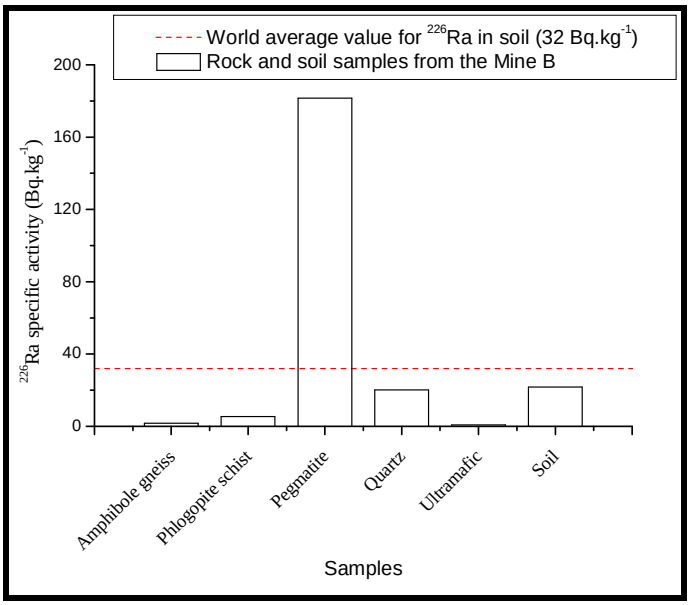

Figure 3: ^{226}Ra specific activities in rocks soil samples from mine B. The comparison between the specific activities obtained and the ^{226}Ra worldwide average in soil.

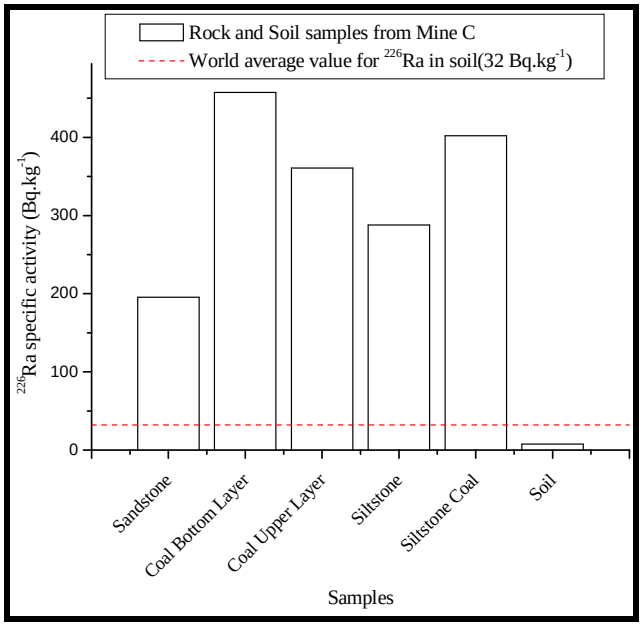

Figure 4: ^{226}Ra specific activities in rock and soil samples from mine C. The comparison between the specific activities obtained and the ^{226}Ra worldwide average in soil.

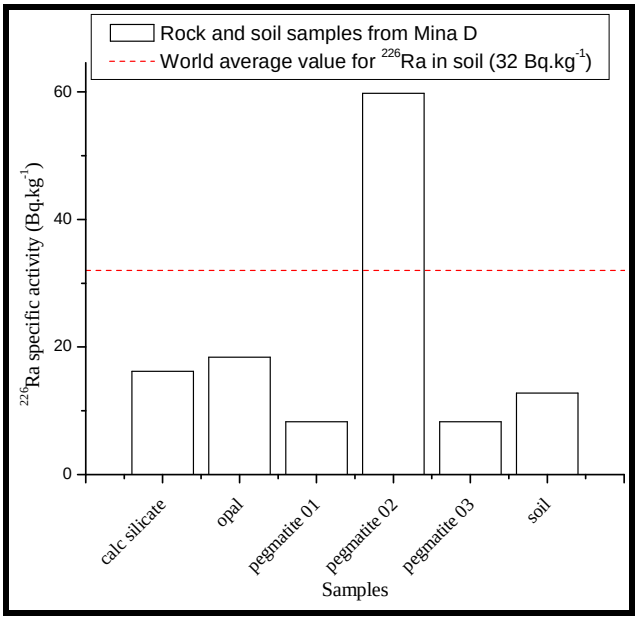

Figure 5: ^{226}Ra specific activities in rock samples and soil from mine D. The comparison between the specific activities obtained and the ^{226}Ra worldwide average in soil.

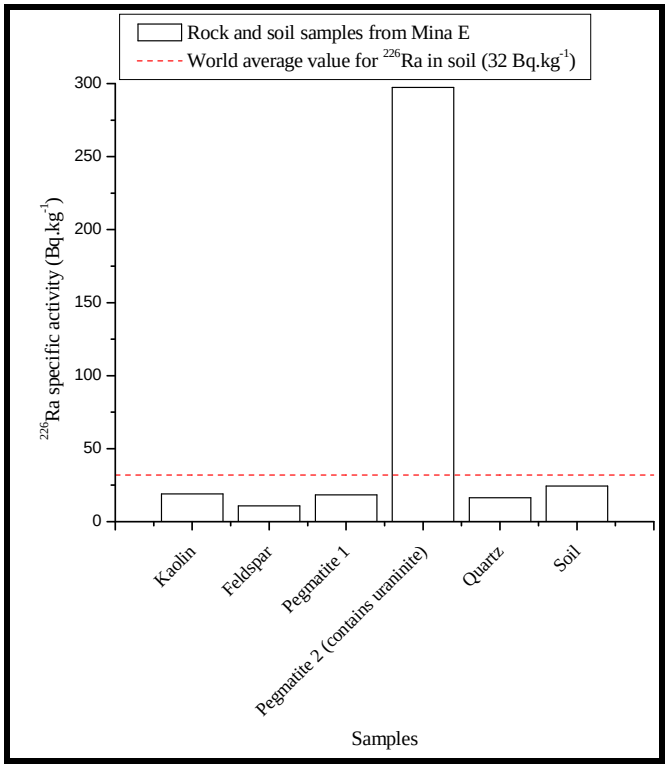

Figure 6: ^{226}Ra specific activities in rock and soil samples from mine E. The comparison between the specific activities obtained and the ^{226}Ra worldwide average in soil.

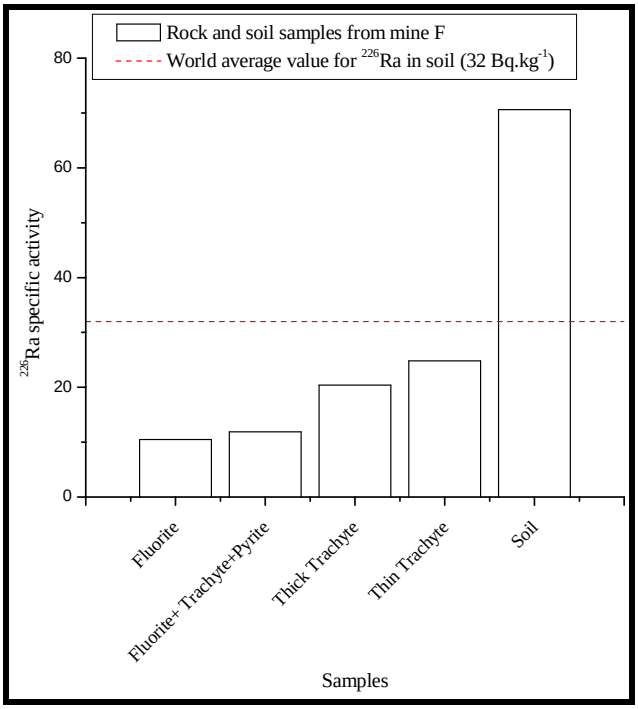

Figure 7: ^{226}Ra specific activities in rock and soil samples from mine F. The comparison between the specific activities obtained and the ^{226}Ra worldwide average in soil.

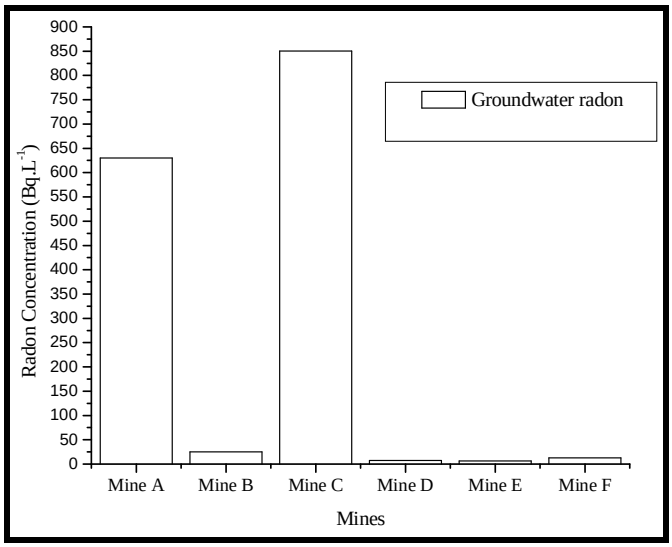

Figure 8: Radon distribution in groundwater in mines surveyed

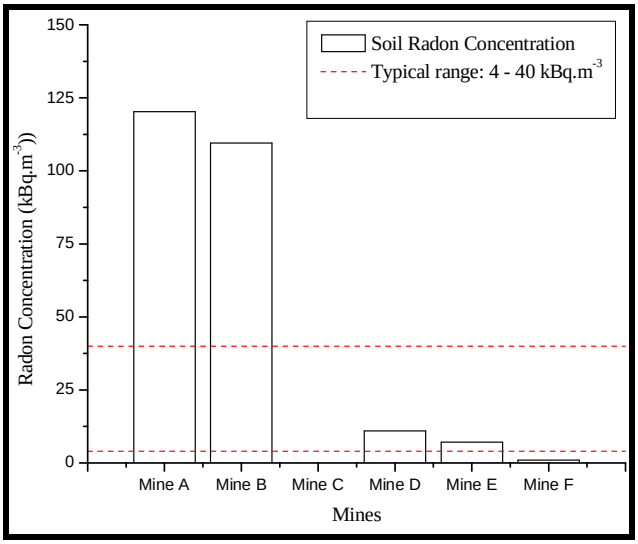

Figure 9: Radon distribution in soil in mines surveyed. The mine C does not have soil radon measurement because the metal probe broke.

3. CONCLUSIONS

According to results shown and discussed above, radon concentration varies considerably from mine to mine and within the same mine. These values are in accordance with the literature. Radon concentration much higher than ICRP actions level have occurred in the all mines, exception mine B and F. Remedial action has to be taken in these mines, for example, by improving mechanical ventilation and appropriate work scale in order to reduce the workers' underground exposure time.

Equilibrium Factors were different in each mine. The mean value was 0.4, range from 0.2-0.7. These results suggest that using a mean value for equilibrium factor to assess dose is not

recommended. The discussion about the radon main source in each specific condition was presented.

Thus, the effective dose estimated for workers of the Brazilian underground mines varies from 1-21 $mSv.a^{-1}$.Only two of the mines (mina A out of operation and mina E) had effective dose above the highest level suggested by ICRP. In addition, the dose effective to miners in these mines exceeds the limit of 20 $mSv.a^{-1}$ for occupationally exposed individual in Brazil.

These results, including the variability, suggest that radon and its progeny measurements should be conducted periodically. It is very important to ensure health and safety standards. Some mines were not investigated; however, they have been part of the Authors' researches. Further studies will also be carried out for a better comprehension of the radioactivity from the radon in the mines. It is important to identify the existence of higher radon concentration points and their causes, to characterize the equilibrium factor and aerosol characteristics for different reference condition of occupational exposure and to assess the corresponding radon risk.

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