Radon exhalation from uranium tailings—comparison of experimental and theoretical data (*)

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Summary. — Two different experimental methods and a theoretical approach are used to calculate ^{222}Rn fluxes in the soil-atmosphere interface. As a common experimental procedure, an open-faced 30 liters cylindrical container was inverted and placed onto the tailing surface with the open face down. After a 24 h period, to balance any possible disturbance, the interior air was analysed. One of the experimental methods is based on integrated ^{222}Rn measurements of the accumulated air-gas mixture, using solid nuclear track detectors placed inside the container. The other method is based on spot air samples using scintillation cells, being the ^{222}Rn concentration measured in equilibrium. The radon flux theoretical values are estimated from the ^{226}Ra content of the tailings material, determined by gamma spectrometry, and using the diffusion equation. The series of results obtained by the referred methods were compared.

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1. – Introduction

Releases of radioactivity associated with uranium mining and milling operations are considered to be a significant source of environmental and radiological problems in and around mining facilities (Busigin *et al.*, 1979).

The Urgeiriça uranium mine is one of the most significant ore deposit in the continental temperate and dry zone of Beira Alta. Exploitation of this mine started in 1913 and it was found to be one of the most important uraniferous deposits in Europe.

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Activities in the mine were entirely dedicated to radium production until 1944, when uranium production became economically interesting. In 1951, a chemical treatment unit for U_3O_8 concentrates was built and in 1967 it was transformed into a new unit with a treatment capacity of about 150 tonnes of ore material per day (Galvão *et al.*, 1988). The mine is now exhausted, but its facilities are still in use for ore treatment transported from other mines.

Milling wastes have been accumulated near the mine and the tailing piles extended over an area of about 11 hectares, with a total deposit amount of 4 million tonnes. The tailing piles, with a high radium content (^{226}Ra activity, in some cases, was found to be above 50 $kBq \cdot kg^{-1}$) could be a major source of ^{222}Rn to atmosphere in the surrounding area.

The main purpose of this study is to assess the ^{222}Rn exhalation rates from Urgeiriça uranium tailings and to compare the results obtained through two different experimental methods and a theoretical approach using the diffusion equation proposed by International Atomic Energy Agency Technical Reports Series.

2. – Materials and methods

Two different tailing piles were selected as sampling locations according to their distinct characteristics and age. The oldest one (T1), impounded to a lower yield processing and ore treatment for uranium recovery, exhibits high ^{226}Ra contents, 47.2 $kBq \cdot kg^{-1}$ on average. The other one (T2) presents an average concentration of 8.1 $kBq \cdot kg^{-1}$.

Regarding the tailings water content, which is a parameter that strongly affects the emanation and diffusion coefficients, and consequently the exhalation rate, significant differences were found. T1 water content remained almost constant during the year, ranging from 27% to 38%, independently of the weather conditions. On the contrary, T2 water content measurements have shown a large variation range, from 1% to 33%, according to rainfall occurrence.

Several open-faced 30 liters cylindrical containers were inverted and placed onto the tailings surface, with the open face down, during 24 h. This period of time was previously tested *in situ*, in order to avoid build-up to a level that could reduce ^{222}Rn concentration by back-diffusion into the measured area (IAEA, 1992).

As ^{222}Rn exhalation rates are influenced by meteorological parameters, several series of measurements were carried out throughout one year, attempting to cover seasonal specific weather conditions.

Two different experimental methods were used for the ^{222}Rn measurements (fig. 1):

Scintillation cells. After a 24 h accumulation period, a sample of the air inside the containers was taken, filtered on a Millipore type AA 0.8 μm filter and collected into a 125 cm^3 scintillation cell. ^{222}Rn activity was measured in equilibrium (after 3 h) on an alpha counter equipment.

Solid track detectors. Integrated measurements were performed using passive nuclear alpha track detectors (LR-115) placed inside the containers and exposed during 24 h. The films were etched and the tracks counted using a spark counter.

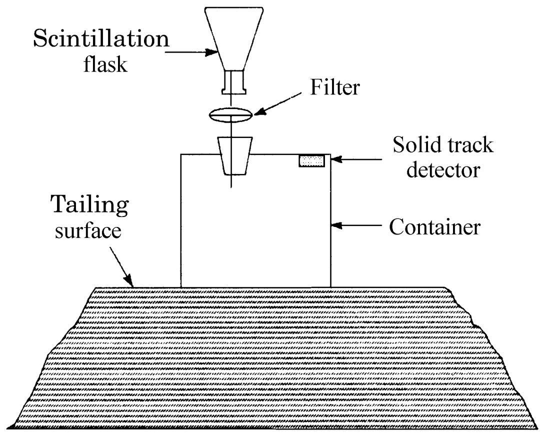

Fig. 1. – Field apparatus showing the two experimental methods.

3. – Theoretical calculations

Samples of the tailings material were collected beneath each container in a 10 cm deep layer, dried at 110° C and their moisture content determined. ^{226}Ra activity was analysed by gamma spectrometry using a Ge(Li) detector and ^{222}Rn exhalation flux calculated through the diffusion equation, as described in the following.

^{222}Rn generation in uranium tailings and its release to atmosphere involve two main mechanisms: escape from particles in which it was formed by recoil movement and diffusion through soil pores until it reaches the atmosphere, where it will suffer dispersion. The ^{222}Rn fraction that will be available to diffusion after release from soil particles is known as the emanation coefficient. ^{222}Rn transport through soil pores is expressed by the diffusion coefficient. The total ^{222}Rn exhalation flux to atmosphere, in $\mathrm{Bq \cdot m^{-2} \cdot s^{-1}}$, can be calculated using the diffusion equation (IAEA, 1992)

$$\phi_{\text{Rn-222}} = R \varrho C_{\mathrm{e}} \sqrt{\lambda C_{\mathrm{d}}}\,, \tag{1}$$

where:

R is the ^{226}Ra activity concentration ($\mathrm{Bq \cdot kg^{-1}}$);
ϱ is the tailing material density ($\mathrm{kg \cdot m^{-3}}$);
C_{e} is the emanation coefficient (adimensional);
λ is the ^{222}Rn decay constant ($2.1 \times 10^{-6} \cdot \mathrm{s^{-1}}$);
C_{d} is the diffusion coefficient ($\mathrm{m^2 \cdot s^{-1}}$).

Equation (1) is valid for uncovered tailings with thickness above 2 meters, for wet tailings, or above 4 meters, for dry tailings (IAEA, 1992). Both tailings here considered lay under these conditions.

To express the material density (ϱ) a value of $1.6 \times 10^3\,\mathrm{kg \cdot m^{-3}}$, typical for tailings (NRC, 1981), was used.

In general the emanation coefficient (C_{e}) ranges from 0.15 to 0.40 (IAEA, 1992), the highest values being related to tailings constituted by lower-size particles. According to previous studies, Urgeiriça tailings emanation coefficients usually range between 0.23 and 0.30 (Reis *et al.*, 1996). An average value of 0.25 was assumed for the present study calculations, which is in agreement with the C_e value recommended by IAEA.

The diffusion coefficient (C_d) is the parameter most affected by the soil water content. A 20% change in the water content value could modify the diffusion coefficient in two orders of magnitude (Rogers and Overmyer, 1980).

In order to adjust the diffusion coefficient according to soil water content effect, the relation proposed by Rogers and Overmyer was applied:

$$C_d = 1.06 \times 10^{-5} e^{-0.261m}, \qquad (2)$$

where m is the soil water content (% by weight).

4. – Results and discussion

Average radon exhalation rate results are presented in fig. 2, for the two tailings under study and during different periods of the year. It can be observed that LR-115 measurement values are significantly lower in comparison with both scintillation cells and theoretical values. The most probable explanation is that condensation inside the containers creates water droplets on the film surface that could act as a barrier for alpha-particles, preventing track formation.

Scintillation cells measurement values for tailing T1 show higher exhalation rates

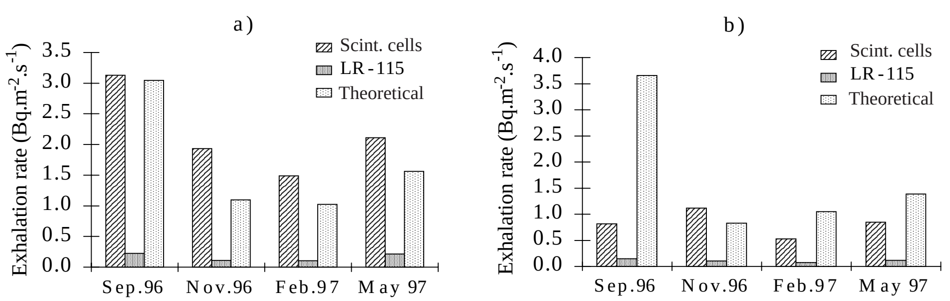

Fig. 2. – Comparison of average exhalation rate values obtained through experimental measurements and theoretical calculations: a) tailing T1; b) tailing T2.

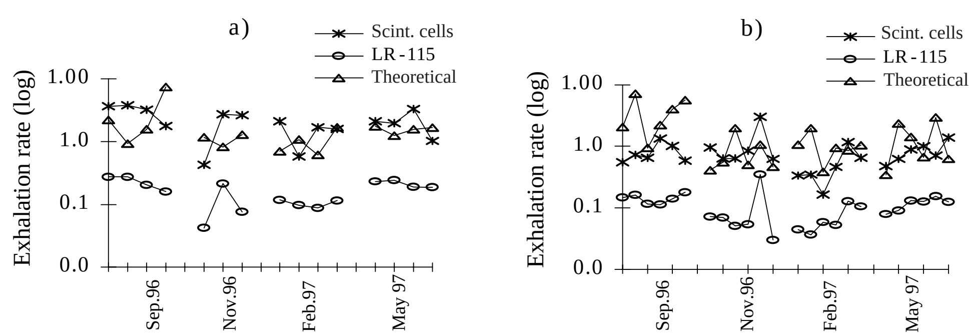

Fig. 3. – Comparison of exhalation rate measurement results for all sampling points in each tailing, during different campaigns: a) tailing T1; b) tailing T2.

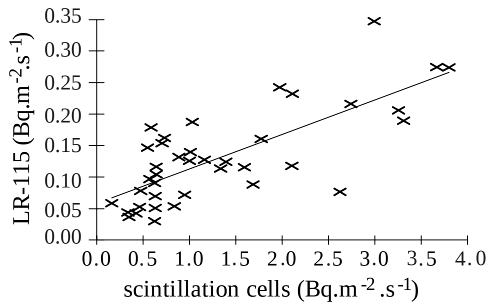

Fig. 4. – Correlation between scintillation cells and LR-115 measurements.

than the calculated ones. An opposite trend is shown for tailing T2, exception made for November values. The reason for this behaviour was the high water content of tailing T2 measured during the November campaign (similar to the ones measured on tailing T1 for all the campaigns). The diffusion coefficients, whose calculation is based on this parameter, have a pronounced decreasing effect on the final exhalation rates calculated through the diffusion equation.

Exhalation rate log values from different sampling points in each tailing are plotted in fig. 3. As already mentioned, LR-115 results are significantly lower than the other two series of values. However a remarkable fact can be drawn from fig. 3. A similar pattern of behaviour is clearly shown in both scintillation cells and LR-115 plots. Theoretical exhalation rates show wider variations, particularly on tailing T2, which was found to be related to pronounced water content changes.

Those results lead us to look up for correlations between the exhalation rates measured through scintillation cells (considered as reference method) and the other two series of results.

A reasonable correlation ($r = 0.7463$) was found between scintillation cells and LR-115 results for both tailings (fig. 4). No correlation was found between scintillation cells and theoretical values.

In an attempt to improve correlation coefficients, regression analysis was calculated for each tailing (fig. 5). The resulting correlation coefficients were not significantly different from the previous ones ($r = 0.6829$ for tailing T1 and $r = 0.7912$ for tailing T2). Once again no correlation was found between scintillation cells and theoretical values.

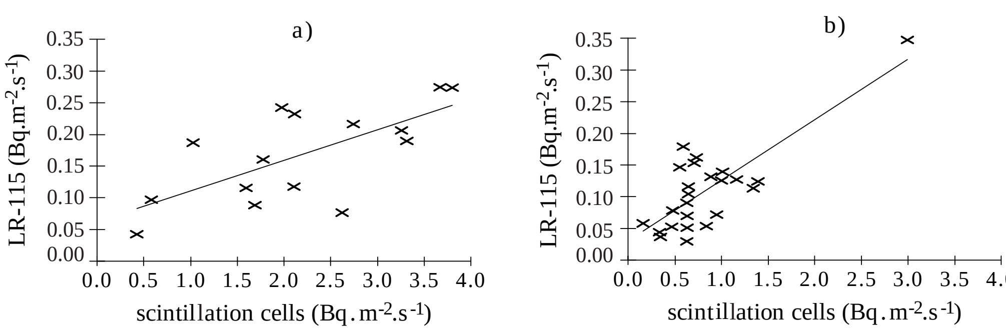

Fig. 5. – Correlation between scintillation cells and LR-115 measurements: a) tailing T1; b) tailing T2.

As mentioned before, the variations on the theoretical exhalation rates are probably related to pronounced changes on soil moisture. This hypothesis is corroborated by the wider variations observed for tailing T2 (fig. 3), where water content values range from 1% to 33%.

Theoretical exhalation rates appear to be very sensitive to the diffusion coefficient (C_d) calculated through eq. (2) where the water content value is estimated on a per cent by weight basis. No significant variation of C_d should occur for moisture contents below 12% (Rogers and Overmyer, 1980). A closer agreement between measured and calculated fluxes would have been found if C_d had been calculated using moisture saturation instead of moisture content (Strong and Levins, 1982). According to the same authors, at low moisture contents water is first absorbed into the internal micro fissures of the particles and has no effect on the diffusion coefficient or on the exhalation flux. When the internal pores become saturated, water fulfils the spaces between particles and the diffusion coefficient is strongly reduced.

5. – Conclusions

According to this study outcomes, passive nuclear alpha track detectors (LR-115) did not produce reliable results. A method using scintillation alpha counting should be employed for field radon exhalation rate measurements.

Theoretical results have shown a high sensitivity to diffusion coefficient. Better results should be obtained if this parameter could be related to the percentage saturation of interstitial void space rather than the percentage moisture by weight.

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