RADON EMANATION MEASUREMENTS FROM URANIUM ORE TAILINGS IN ARGENTINA

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The uranium ore tailings represent important release sources of radon gas (Rn-222) into the atmosphere, being one of the main concerns regarding the environment in the vicinity of uranium mining and milling areas. The tailings area remaining after the mill has ceased operation becomes a long-term source of environmental radioactive contamination due to wind or water erosion and mainly to radon emanation.

Radon gas, with a radioactive half-life of 3.8 days, emanates from the ore tailings as a radioactive decay product of radium-226 (Ra-226, 1600 years), which is generated by the decay of thorium-230 (Th-230), with 8.10^4 years of half-life. Emanation from the tailings arises primarily from radon produced in the surface layer, and up to about 20 % diffuses out of the crystallized structure of the mineral (1). The Rn-222 emanation rate depends on many characteristics, including parents content in the tailings, ore particle size, moisture content, porosity and mineral species (2). Besides it, the gaseous diffusion may be influenced by meteorological factors such as atmospheric pressure, temperature and humidity.

Since 1983, a field team from the Environmental Radioactivity Section of CNEA, comprising the authors of this report, has carried out measurements of the emanation rates of Rn-222 from uranium mining and milling tailings located at different regions in Argentina, with the aim of estimating the resulting radiological impact.

RADON EMANATION RATE MEASUREMENTS

The Rn-222 emanation measurements were made by using the technique develloped by R.J. Countess (3). In this method, cilindric canisters containing activated charcoal are positioned on the tailing surface for a known period, to collect radon released from it. After collection, the canisters are stored for at least three hours to allow Bi-214, a gamma-ray emitting daughter, to grow towards equilibrium with the parent radon. The Bi-214 activity is counted through the 609 keV gamma emission because of its clear separation from other emission lines and the low background in such spectral area. The counting is carried out inside a lead-brick shield, with a 4 x 5-in. NaI crystal detector coupled to a multichannel pulse-height analyzer.
Rn-222 emanation rate is estimated taking into account the measured Bi-214 activity corrected for decay during the storage period, the sampling time and the exposed surface.
Assuming a charcoal adsorption efficiency similar to that reported by other authors,(4,5), a lower detection limit of 0.15 $Bq.m^{-2}.s^{-1}$ is obtained for the experimental conditions, with a statistical error of 10%.

RADIUM-226 CONTENT OF TAILINGS

The radium content of the sterile material beneath each site of emanation measurement was determined. Samples were taken from under the radon collectors located at each site, dried to determine the moisture content, homogenized and packed in small plastic bottles, sealed, and stored for allowing them to reach radioactive equilibrium. Afterwards, the samples were placed into a Ge(Li) spectrometer for analysis. Ra-226 content was estimated by using the data from Pb-214 and Bi-214 peaks in the resulting gamma spectra.

DOSE ASSESSMENTS

The uranium mining and milling sites are usually located at low population density areas. This is the case for all the reported areas but one, Malargue, which is a milling installation situated near from the homonymous city (about 500 m).
The individual and collective dose commitments from radon releases were estimated using the methodology presented by UNSCEAR (1). For the annual effective dose equivalent assessments to the most exposed members of the public, the model used assumes an atmospheric dilution factor of 5.10^{-6} s m^{-3} at 500 m from the sources, where the critical groups are supposed to live.
The collective effective dose equivalent commitments from these tailings were estimated by using a simplified model, taking into account the natural radon emanation from soil and the corresponding radon concentration in air. These collective doses were calculated over the mean life of Th-230 (1.1 10^{5} y).

RESULTS

Measurements of radon emanation rate and radium content were made at four sites in Argentine uranium mining and milling areas. For each site, the corresponding distributions of Rn-222 emanation rates are shown in Figs. 1 to 4.
In Table 1, the Ra-226 concentrations are compared to the corresponding Rn-222 emanation rates.

TABLE 1. Rn-222 emanation rates

SITE	IMPOUNDMENT AREA (ha)	Rn-222 EMANATION RATE (Bq/m2 s)	Ra-226 CONC. (Bq/g)	RATIO
S.Rafael	5.0	7.7	8.1	0.95
Malargue	3.0	5.8	8.1	0.71
Salta	7.5	20.6	9.4	2.19
Chubut	7.0	2.7	4.9	0.56

The calculated doses to most exposed members of the public, expressed as annual effective dose equivalents, and the collective effective dose equivalent commitment for each site are listed in Table 2.

TABLE 2. Individual and collective dose commitments

SITE	Rn-222 RELEASE TBq/a	INDIV.DOSE uSv/a	COLL. DOSE man Sv	(NORMALIZED) man Sv/GW(e).a
S.Rafael	12.0	108.0	8.6E+3	3.0E+3
Malargue	5.5	49.6	4.0E+3	7.0E+2
Salta	49.0	441.2	3.5E+4	1.0E+4
Chubut	6.0	54.0	4.3E+3	3.6E+3

CONCLUDING REMARKS

The calculated specific radon emanation rate, the ratio of the radon emanation rate to Ra-226 concentration, varied from site to site, ranging from 0.56 to 2.19 Bq m^{-2} s^{-1} per Bq g^{-1} (Table 1). These values are compatible with those published by UNSCEAR, having into account that the values depend on meteorological conditions, such as wind speed, atmospheric stability and rainfall (1). Other factors that may affect this ratio are the spacial distribution of radium, and radon transport parameters, particularly moisture (5).

The above expressed explains the difference observed in the specific radon emanation rates for San Rafael and Malargue tailings where both radium concentrations are similar.

In the case of Salta, where the ratio is the highest, geographical and meteorological factors contribute to increase the radon emanation rate.

The estimated doses are in agreement with UNSCEAR reports. The individual doses to critical groups represent only a few percent of overexposure to natural radiation.

Collective dose commitments are extremely uncertain and highly speculative because of the assumptions of the duration of the constant release and of the fixed population density and habits. Furthermore, present day tailings managements may lead to radon emanation rates lower than the ambient levels for soils in the mill vicinity,so that almost no long term dose commitment arises.

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Fig. 1

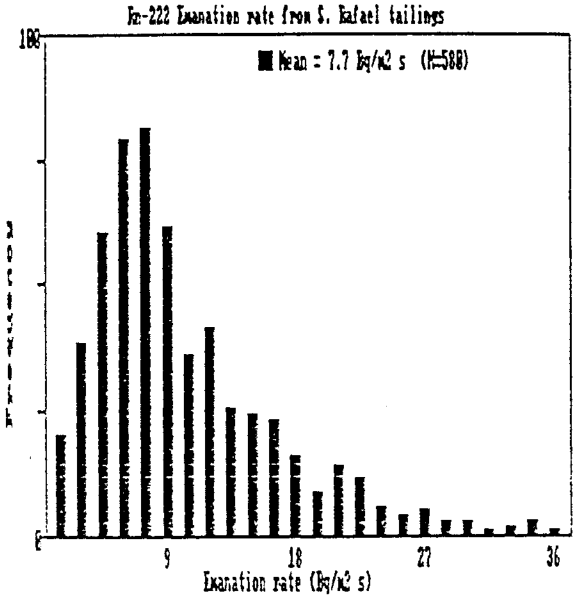

Fig. 2

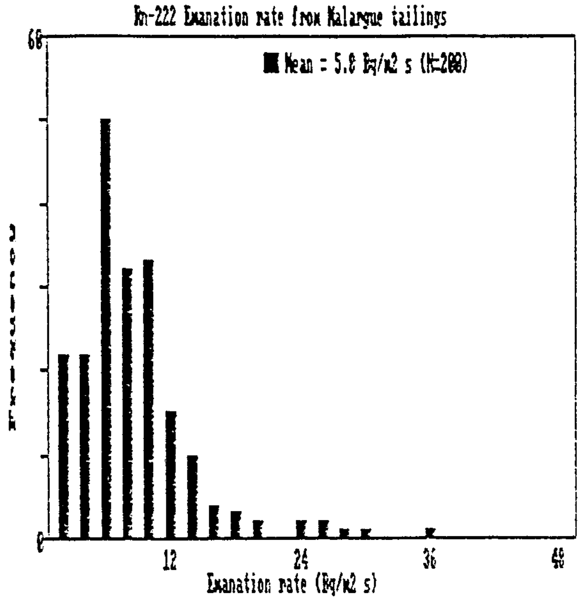

Fig. 3

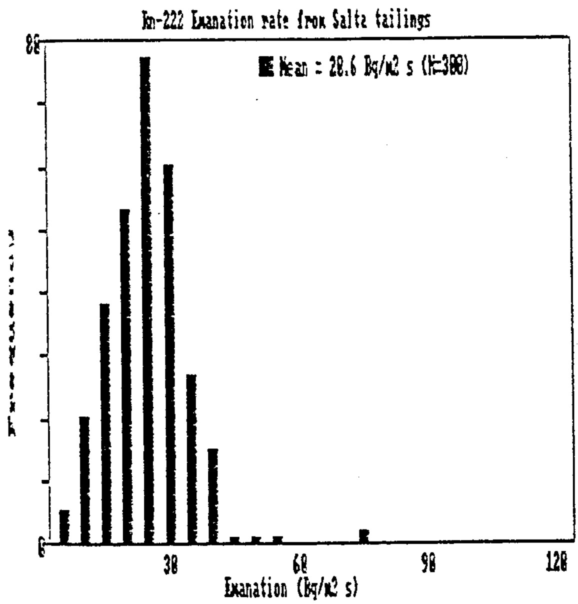

Fig. 4