Measurements of ^{222}Rn , ^{220}Rn and Their Decay Products in the Environmental Air of the Errachidia Area (Morocco) Using SSNTDs

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Abstract Alpha and beta activities per unit volume of air, due to radon, thoron and their decay products were measured in the outdoor atmosphere in Errachidia city, Morocco, by using both CR-39 and LR-115 type II solid-state nuclear track detectors (SSNTDs). Simultaneously, meteorological parameters such as air temperature, atmospheric pressure, and relative humidity were also measured in situ. In addition, the outdoor radon activities show the highest value of the outdoor radon activity was determined in the early morning hours, and the lowest values were found in the afternoon, over a three month study period, between April-June 2006. The diurnal variations in the outdoor radon activity are found to exhibit correlation with relative humidity and negatively *correlate* with the air temperature. The annual effective dose in the present study for Errachidia environment is found to be 0.29 mSv/y.

Keywords Nuclear Track Detectors; Radon; Thoron; Environment; Dose Assessment.

1. Introduction

The continuous exposure of human beings to ionizing radiation from natural sources is inevitable on Earth. The total amount of natural radiation exceeds that released from all the man made sources combined. Corresponding to the UNSCEAR recommendation [1], the Radon isotope (^{222}Rn) is the most important source of natural radiation and is responsible for approximately 45 % of the effective equivalent dose to man, released by natural radioactive sources, while the rest is due to high energy cosmic rays, terrestrial gamma rays and radionuclides in the body, except radon [1]. The radionuclides radon and thoron are produced

through decay of ^{226}Ra and ^{224}Ra in the earth's crust. These gases are free to move through soil pores and rock fractures, and then to escape into the atmosphere. Once in the atmosphere, the ^{222}Rn and ^{220}Rn nuclei decay, producing isotopes of polonium, lead and bismuth, and also thallium from ^{220}Rn . The ^{220}Rn relatively short-half life (56.6 s) and does not have much time to travel from its production site to the immediate environment of human beings.

The variation of concentration of the progeny of radon isotopes in air depends on the place, time and height above the ground, and on meteorological conditions, such as temperature, relative humidity and wind speed.

In this work, the measurement method is based on using CR-39 and LR-115 type II solid state nuclear track detectors (SSNTDs) for measuring the concentration of radon, thoron and their progenies in the outdoor air near the university city of Errachidia in Morocco. The atmospheric radioactivity was monitored for three months, during April, May and June 2006. Errachidia town is located in the Meknes-Tafelalet region (31°55'55" N and 4°25'28" W), at an altitude of 1029 m. The measurements were performed at a fixed site, at 1.7 m above the ground, for a period of 90 days, three times per day at the following times: 4:00-10:00 (morning), 13:00-18:00 (midday), and 18:00-23:00 (evening). We also determined effective equivalent doses due to the radon progeny to the members of the public.

2. Materials and Methods

Disc shaped Pershore Mouldings CR-39 (500 μm thickness) and Kodak LR-115 type II (12 μm cellulose nitrate on 100 μm polyester base) solid state nuclear track detectors (SSNTDs) of radius $q = 2$ cm have been placed in the outdoor atmosphere (Figure 1) for five hours. After the irradiation, the exposed films were etched in two NaOH solutions at optimal conditions of etching, ensuring good

sensitivities of the SSNTDs and a good reproducibility of the registered track density rates: the first solution of 2.5 mol L⁻¹ normality at 60°C for 120 minutes for the LR-115 II films and the second solution of 6.25 mol L⁻¹ normality at 70°C for 7 hours for the CR-39 detectors [2]. After this etching treatment, the track densities registered on the CR-39 and LR-115 II SSNTDs were determined by means of an optical microscope with 40x magnifying power. For our experimental etching conditions, the residual thickness of the LR-115 type II SSNTD is 5 µm which corresponds to the lower ($E_{\min}=1.6$ MeV) and upper ($E_{\max}=4.7$ MeV) energy limits for the registration of tracks of alpha-particles in the LR-115 II films [3]. All α -particles emitted by the radon and thoron series that reach the LR-115 II detector surface under an angle lower than its critical angle of etching, θ'_c with a residual energy between 1.6 MeV and 4.7 MeV are registered as bright track-holes. The CR-39 detector is sensitive to all α -particles reaching its surface under an angle smaller than its critical angle of etching θ_c . The angles of etching θ'_c and θ_c were calculated by using a method described in detail by Misdaq et al [4].

Figure 1. Arrangement of the solid state nuclear track detectors of radius $q=2$ cm placed in outdoor air. The distance between the SSNTDs and the ground level is of 1.7m.

The global track density rates (tracks cm⁻² s⁻¹), due to α -particles emitted by the radon (three α -emitters) and thoron (four α -emitters) series within a volume of air at an outdoor location, registered on the CR-39 (ρ_G^{CR}) and LR-115 II (ρ_G^{LR}) detectors, after subtracting the corresponding backgrounds, are given by [5]:

$$\rho_G^{CR} = 0.25 \left[\sum_{j=1}^3 \sin^2(\theta_c^{CR}) A_j K_j R_j + \sum_{j=1}^4 \sin^2(\theta'_c)^{CR} A_j K'_j R'_j \right] \quad (1)$$

and

$$\rho_G^{LR} = 0.25 \Delta R \sin^2(\theta_c^{LR}) \left[\sum_{j=1}^3 A_j K_j + \sum_{j=1}^4 A_j K'_j \right] \quad (2)$$

Where A_j (Bq.cm⁻³) is the α -activity of the j^{th} α -emitter, R_j and R'_j are the ranges in air of an α -particle of index j and initial energy E_j emitted by the nuclei of the radon and thoron series in the outdoor air of a location, respectively, K_j and K'_j are respectively the branching ratios corresponding to the disintegration of the nuclei of the radon and thoron groups and $\Delta R = R_{\max} - R_{\min}$. $R_{\min}$ and $R_{\max}$ are the α particles ranges in air which correspond to the lower ($E_{\min}$) and upper ($E_{\max}$) ends of the energy window ΔE . The ranges of the alpha-particles emitted by the radon and thoron series in air were calculated by using a TRIM program [6].

Radon and thoron formed in the soil are released into the atmosphere. After this exhalation, the radioactive gases and their daughters are distributed in outdoor air by means of diffusion. The volumic radioactivity is influenced by the decay constant of the radionuclide, and by the wet and dry removal of aerosols from the atmosphere.

Neglecting the wet removal and taking into account a uniform exhalation rate of radon/thoron in the x, y -plane (no horizontal gradient), and assuming steady-state conditions above the ground level ($z=0$), the activities of a $(j-1)^{\text{th}}$ nucleus A_{j-1} and its j^{th} A_j decay product of the radon (^{222}Rn ($j=0$), ^{218}Po ($j=1$), ^{214}Pb ($j=2$), ^{214}Bi ($j=3$) and ^{214}Po ($j=4$) and thoron (^{220}Rn ($j=0$), ^{216}Po ($j=1$), ^{212}Pb ($j=2$), ^{212}Bi ($j=3$) and ^{212}P ($j=4$)) series are related by [7]:

$$\frac{D_T}{\lambda_j} \frac{\partial^2}{\partial z^2} A_j(z) + A_{j-1}(z) - A_j(z) = 0 \quad (3)$$

Where D_T is the turbulent diffusion coefficient of the outdoor air atoms and λ_j is the decay constant of the j^{th} radionuclide.

Solutions of Equation 3 are obtained under the following boundary conditions:

$$A_j(z=0) = 0 \quad \text{for } j=1,2,3 \text{ and } 4, \quad \text{and} \\ A_j(z \rightarrow +\infty) = 0 \quad \text{for } j=0,1,2,3 \text{ and } 4 \text{ that is, far from the ground.}$$

By measuring the ρ_G^{CR} and ρ_G^{LR} track density rates and combining equations 1-3 one can evaluate the concentrations of radon and its decay products and thoron and its progeny in the outdoor air of a location by using a Maple 8 code [8].

The annual effective dose (mSv/y/h of exposure) due to the radon progeny to the members of the public was also estimated according to the following formula [1]:

$$E = A_c(^{222}\text{Rn}) \cdot F \cdot t \cdot D \quad (4)$$

where $A_c(^{222}\text{Rn})$ is the radon activity or radon concentration in the outdoor air of a location (Bq/m³), $F=0.6$ is the equilibrium factor between radon and its progeny in outdoor air, $t = 1$ h/y and $D = 9.0 \cdot 10^{-6}$ mSv (Bq/m³ h)⁻¹ is the dose conversion factor [1].

Table 1. Data obtained for the alpha and beta activities per unit volume due to radon, thoron and their decay products in the outdoor air for Errachidia town over the three months period (April-June 2006).

Parameter	Temperature (C°)	Relative humidity (%)	Radon decay products				Thoron decay products			
			$A_c(^{222}\text{Rn})$ (Bq m ⁻³)	$A_c(^{218}\text{Po})$ (Bq m ⁻³)	$A_c(^{214}\text{Pb})$ (Bq m ⁻³)	$A_c(^{214}\text{Po})$ (Bq m ⁻³)	$A_c(^{220}\text{Rn})$ (Bq m ⁻³)	$A_c(^{216}\text{Po})$ (Bq m ⁻³)	$A_c(^{212}\text{Pb})$ (Bq m ⁻³)	$A_c(^{212}\text{Po})$ (Bq m ⁻³)
Minimum	10	10	4.35±0.38	4.02±0.35	2.34±0.2	1.76±0.15	0.34±0.01	0.34±0.01	0.054±0.004	0.048±0.004
Maximum	40	90	18.08±1.29	17.00±1.36	9.75±0.69	7.23±0.52	1.71±0.13	1.71±0.13	0.281±0.021	0.23±0.017
Average	25	36	8.84±0.72	7.95±0.68	4.68±0.42	3.45±0.28	0.68±0.05	0.68±0.05	0.1±0.008	0.095±0.008
Number of results	90	90	270	270	270	270	270	270	270	270

Table 2. Results for outdoor radon activity and radon effective dose in morning, afternoon and evening over the three months period (April-Jun, 2006)

Parameter	Morning		Afternoon		Evening		Effective dose (mSv y ⁻¹)
	$A_c(^{222}\text{Rn})$ (Bq m ⁻³)	$A_c(^{220}\text{Rn})$ (Bq m ⁻³)	$A_c(^{222}\text{Rn})$ (Bq m ⁻³)	$A_c(^{220}\text{Rn})$ (Bq m ⁻³)	$A_c(^{222}\text{Rn})$ (Bq m ⁻³)	$A_c(^{220}\text{Rn})$ (Bq m ⁻³)	
Minimum	4.35±0.38	0.34±0.01	4.15±0.3	0.32±0.03	4.00±0.3	0.3±0.03	0.068
Maximum	18.08±1.29	1.71±0.13	11.25±0.8	1.68±0.14	9.85±0.8	1.5±0.13	0.29
Average	8.84±0.72	0.68±0.05	8.10±0.67	0.6±0.05	8.6±0.75	0.55±0.05	0.14
Number of results	180		180		180		270

3. Results and Discussion

The alpha and beta activities due to radon, thoron and their decay products, per unit volume of air in the outdoor atmosphere, in areas frequented by students, were measured over a period of three months between April-June 2006, and the results are reported in Tables 1 and 2. At the site, the meteorological parameters: wind speed, air temperature, air pressure and relative air humidity were obtained using commercially available sensors and the climate program WeatherLink 4.04f (Figure 2) [9]. Since the track detectors utilized were etched in two NaOH solutions at optimal conditions of etching, ensuring good sensitivities of the SSNTDs and good reproducibility of the registered track density rates, their backgrounds were determined and subtracted from the measured global track density rates registered on these films in the location studied. They were determined by means of the same optical microscope with magnifying power 40x. Though, the statistical uncertainty is predominant in the track counting. Indeed, from the statistical error on track counting one can determine the error on the track density rate and then evaluate the relative uncertainty of the determination for radon, thoron and their progenies, which is 10 %. One notes that alpha and beta activities due to radon and its progeny are higher than those due to thoron and its daughters for the outdoor air studied. Indeed, due to its too short half-life (55s) thoron (^{220}Rn) has a diffusion length in air smaller than that of radon (^{222}Rn), which has a longer half-life (3.82 d). A secular radioactive equilibrium is established between ^{214}Bi (19.9 min) and ^{214}Po (165.10^{-6} s) nuclides of the radon series. A secular radioactive equilibrium is established between ^{220}Rn (55s) and ^{216}Po (0.15 s) and between ^{212}Bi (1 h) and ^{212}Po ($0.3.10^{-6}$ s) nuclides of the thoron series.

s) nuclides of the thoron series.

As shown in Table 1, Alpha and beta activities per unit volume due to radon varied within the range (4.35 ± 0.38) Bq.m^{-3} and (18.08 ± 1.29) Bq.m^{-3} with an average value of (8.84 ± 0.72) Bq.m^{-3} . A typical behavior with high outdoor radon activity in the early morning and low outdoor radon activity in the afternoon was observed over the 3 months period of investigations, as can be seen in Table 2. The outdoor radon activity averages corresponding to the different sessions are 8.84 ± 0.72 , 8.10 ± 0.67 and 8.6 ± 0.75 Bq.m^{-3} for morning, afternoon and evening, respectively. Data obtained are in good agreement with each other authors [10-15]. It can be explained mainly by the variation of the atmospheric stability affected by the time variations and spatial differences in the temperature of the ground and surface air as a result of the solar radiation flux. There are regular periodic changes in the atmospheric stability that are caused by the solar heating of the soil surface which in turn causes heating of the near-surface atmosphere. During the day, the temperature increases and temperature differences come into being due to solar radiation. The surface air circulates in a thermoconvective motion and therefore radon and radon progeny get dispersed vertically in a thick air layer. After sunset the surface air layer cools down and temperature differences decrease, which cause an increase in the atmospheric stability (thermoconvective motion weakens); so radon and radon progeny accumulate near the surface. Since the temperature reaches its maximum in the afternoon and minimum in the early morning, radon and radon progeny concentrations are low in the afternoon and high in the early morning. [16-19].

Figure 2. WeatherLink illustration

Figure 3. Correlation between outdoor radon activity and temperature air.

Figure 4. Correlation between outdoor radon activity and relative humidity.

Figure 3 shows the negative correlation between temperature and outdoor radon activity, which is approximately exponential with a correlation coefficient of $R^2 = 0.75$. As shown in Figure 4, the correlation between relative humidity and outdoor radon activity is also approximately exponential but positive, with a correlation coefficient of $R^2 = 0.69$. This is due to the fact that as temperature increases, the relative humidity decrease resulting in the decrease of moisture content in the atmosphere [20].

The annual effective dose due to the radon progeny to the members of the public was also estimated according to the UNSCEAR formula (equation 4). For individuals spending 8 h per day (hence, 2920 h/y) in outdoor atmosphere of

Errachidia, the maximum effective dose due to radon progeny was found to be 0.28 mSv/y, a value lower than the 1.15 mSv/y committed effective dose limit given by UNSCEAR [1].

5. Conclusion

The study showed that by using both C-39 and LR-115 type II SSNTDs one can evaluate the concentration of radon, thoron and their progenies in the outdoor atmosphere. The measured atmospheric concentration of radon and its progeny in Errachidia city reach a maximum value in the afternoon.

The stable atmosphere during the night helps more accumulation of radon and hence, the night time concentrations are higher than the daytime atmospheric concentrations of radon.

The concentrations are the lowest in the afternoon, when the atmosphere is unstable. A correlation between radon concentration and relative humidity, and the negative correlation with temperature air is observed. The annual effective dose found in the present study in the atmosphere of Errachidia town is of 0.29 mSv/y which is smaller than the "normal" background level of 1.1 mSv/y; as quoted by UNSCEAR [1] and less than the maximum permissible dose defined by the International Atomic Energy Agency (IAEA) which is about 5 mSv/y [21]. This SSNTD technique, which has the advantage of being inexpensive, sensitive and accurate, and does not need the use of any radon standard source for its calibration, is a good tool for assessing the radiation dose risk due to the inhalation of outdoor air.

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