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A mathematical description of the diurnal variation of radon progeny

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Abstract

The variation of the alpha (α) radioactivity in the air near the ground and the ground-level total gamma (γ) radiation has been monitored in North-eastern Greece over several days. Meteorological information regarding the temperature of the air and humidity has been simultaneously recorded. The α-radioactivity shows a periodic diurnal variation with a peak in the morning followed by a decrease in the afternoon; then, the variation rises again to the peak the next morning. The variation of the ground γ-radiation follows that of the air α-radioactivity. Furthermore, their significant dependence on the air temperature and humidity is confirmed, rising with an increase in humidity and a decrease in temperature. Hence, a mathematical function has been developed to describe the diurnal variation of the α-radioactivity in terms of the ground-level γ-radiation and the meteorological variables of temperature and humidity.

Keywords: Alpha air radioactivity; Radon progeny; Ground gamma radiation; Radon progeny; Modelling

1. Introduction

Natural radioactivity background is a parameter that characterises a particular area comprising the α-radioactivity in the air and the γ-radiation from the ground. Both are due to the radon released from the primordial radionuclides ^{238}U and ^{232}Th in the ground (Venuti et al., 1990; Porstendorfer, 1994), while the γ-radiation further originates from ^{40}K. Continuous monitoring of the α-radioactivity may be effectively used for the timely detection of its sudden unexpected increase above the background level, giving rise to a possible nuclear emergency for the area (Ermilov and Yaryna, 1989). The increase may be due to accidental or operational releases of α-radioactivity into the atmosphere. The former refers to the case of fallout from an accident in a nuclear installation. The latter includes the releases from an installation due to routine operations or indeed undeclared-clandestine-activities within the installation.

Early warning of nuclear emergencies requires real time monitoring of the natural α-radioactivity in the air. Then, information on the temporal variation of the radioactivity in the lower layers of air is obtained. This variation correlates with air temperature, humidity, air speed, atmospheric electric field and air flow direction (Garcia-Talavera et al., 2001; Latha, 2003; Sesana et al., 2003).

In this work, a mathematical function is presented which describes the trend of the diurnal variation of α-radioactivity in the air near the ground. The function is used to normalise the measured variation in a way to enhance the measurement of sudden unexpected peaks of low-level radioactivity over the natural background. The work is based on the monitoring of the diurnal variation of α-radioactivity in the air near the ground, the γ-radiation at ground level and the environmental variables of relative humidity and temperature. Then, the mathematical function developed relates the α-radioactivity with the γ-radiation and these environmental variables.

2. Materials and methods

Radon from the primordial radionuclides ^{238}U and ^{232}Th in the ground is transported through the subsoil pore spaces and released into the atmosphere. The amount released depends on the amounts of ^{238}U and ^{232}Th in the ground, as well as the porosity, dampness and temperature

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of the soil cover. Radon and its progeny are transported, by diffusion and advection, from the ground pore spaces to soil and finally to air as the radionuclides drift in the form of aerosols. Radon (^{222}Rn) from the ^{238}U series decays with the emission of α-particles and has a long enough half-life (3.82 days) in order to diffuse out of the soil and into the atmosphere where it will remain for several days. Among the daughters of radon, ^{214}Pb and ^{214}Bi are significant emitters of γ-radiation with energies at 242 keV (8%), 295.2 keV (18%), 352 keV (35%) the former and 609 keV (43%), 768 keV (5%), 934 keV (3%), 1120 keV (14%), 1238 keV (6%), 1377 keV (7%), 1764 keV (16%), 2204 keV (5%) the latter.

The α-radioactivity in the air near the ground, from the short-lived radon progeny ^{218}Po, ^{214}Pb and ^{214}Bi, the total γ-radiation at ground level in the energy range 0.040–3 MeV and the meteorological variables were measured simultaneously by the Nuclear Technology Laboratory of Democritus, University of Thrace, in a flat area of the city of Xanthi in North-eastern Greece (Seftelis et al., 1995).

The measurement of α-radioactivity (Bq/m^3) was carried out every 10 min and was based on the percolation of air through a tape type paper filter having a pore diameter of 0.5 Å (FAG, 1989). The air inlet to the measurement device was 12 m above the ground. The radioactivity was measured using a 2 in. thick plastic-type detector coated with ZnS (50 mm diameter) positioned right on the filter. The γ-radiation (kcpm) was measured every 10 min, using a 3 in. × 3 in. NaI(Tl) detector. The air temperature (K) and relative humidity (%) were simultaneously monitored using commercial sensors. Radioactivity measurements and meteorological data were stored on a personal computer for further processing.

3. Results and discussion

The simultaneous measurements of the α-radioactivity, γ-radiation and environmental variables are presented for a typical day. Such a day is characterised by the periodic variation of radioactivity rising in the morning and decreasing in the afternoon, a situation encountered in days having a wind speed less than 4 m/s and no precipitation.

The variation of α-radioactivity in the air near the ground is shown in Fig. 1 over a demonstrated period of 5 consecutive typical days in the month of September. The error associated with the measurement of the α-radioactivity from the radon progeny is ±22.5% (Smesters, 1995). The periodic form encountered in the variation of α-radioactivity is consistent with other studies (Porstendorfer et al., 1991; Sesana et al., 2003). The differences in the diurnal variation between the 5 days are due to the different meteorological variables. The minimum and maximum values encountered in the diurnal variation of α-radioactivity differ by factors ranging between 3.5 and 7. The measured total γ-radiation from the ground was in the range 1.160–1.245 kcpm with an average error of ±2.9%.

It was already mentioned that the increase of the α-radioactivity in the air near the ground is expected to be accompanied by an increase in the γ-radiation at ground level. This stems from the fact that some of the radon progeny emit, besides α particles, significant γ-radiation as well. The variation of the γ-radiation is shown in Fig. 1 over the period demonstrated. The trend of the variation follows that of the α-radioactivity, with a peak in the morning and a lower value in the afternoon, which then rises to the following morning.

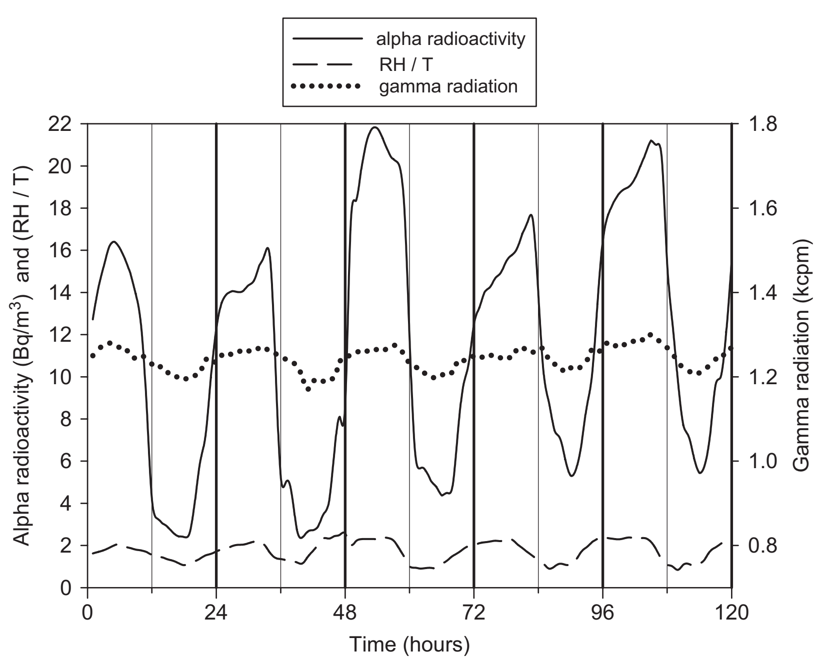

Fig. 1. Variation of measured radioactivity and meteorological variables over 5 consecutive days (120 h).

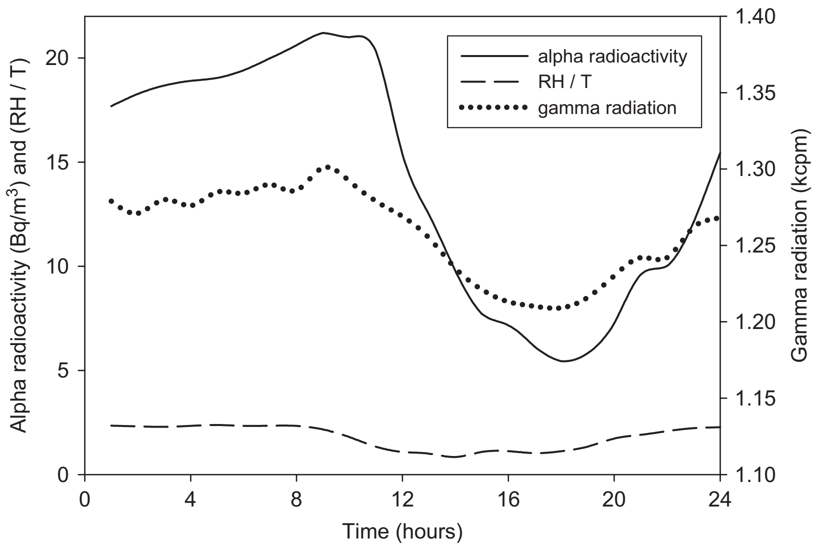

Fig. 2. Variation of measured radioactivity and meteorological variables over 24 h.

The diurnal variation of α-radioactivity in the air has been found to be proportional and inversely proportional to the relative humidity (RH) and temperature (T), respectively (Seftelis et al., 2007). The variation of the ratio of relative humidity and temperature (RH/T) is shown in Fig. 1 over the demonstrated period of the measurements and its trend is similar to that of the α-radioactivity. The changes in the meteorological variables precede those of the radioactivity, since the eventually measured α-radioactivity lags in time behind any changes in meteorological conditions. The detailed analysis of the data from the demonstrated period revealed that, in relation to the α-radioactivity and γ-radiation: (1) the decrease in (RH/T) in the morning occurs 2–3 h and 1–2 h earlier, respectively; (2) in the afternoon, the increase in (RH/T) occurs 1–2 h earlier. The variation of radioactivity and meteorological variables over 24 h is shown in the case of the third day of the demonstrated period of the measurements (Fig. 2). The trend of the variations of α-radioactivity and γ-radiation follows that of (RH/T).

A mathematical function was developed to describe the trend of the diurnal variation of the α-radioactivity in the air near the ground during a typical day. The model is based on the measured α-radioactivity in the air near the ground, γ-radiation at ground level, relative humidity and temperature. The development of the model was based on the hypothesis

$$\alpha = f(\mathrm{RH}, T, \Delta\gamma), \tag{1}$$

where α (Bq/m^3) is the α-radioactivity in the air near the ground, $\Delta\gamma$ (kcpm) is the change in γ-radiation at ground level, RH (%) is the relative humidity and T (K) is the temperature. The parameter $\Delta\gamma$ is defined as the difference between the γ-radiation at ground level (γ) and the minimum value of γ encountered in the measurements over a 24 h period ($\gamma_{\min}$), that is $\Delta\gamma = \gamma - \gamma_{\min}$.

Since the trend of the diurnal variation of the α-radioactivity in air is similar to the ratio of relative humidity and temperature (Fig. 1), the hypothesis is expected to take the form

$$\alpha = f\left(\frac{\mathrm{RH}}{T}, \Delta\gamma\right) \tag{2}$$

or

$$\alpha = f_1\left(\frac{\mathrm{RH}}{T}\right) f_2(\Delta\gamma). \tag{3}$$

Then, on the assumption that function f_1 is linear,

$$\alpha = k\left(\frac{\mathrm{RH}}{T}\right) f_2(\Delta\gamma). \tag{4}$$

On the basis that the radioactivity concentration follows the natural exponential law of radioactive decay, factor f_2 is assumed to have the form

$$f_2(\Delta\gamma) = \mathrm{e}^{\mu\,\Delta\gamma} \tag{5}$$

and hence,

$$\alpha = k\left(\frac{\mathrm{RH}}{T}\right) \mathrm{e}^{\mu\,\Delta\gamma}, \tag{6}$$

where k (Bq K/m^3) and μ (kcpm^{-1}) are coefficients that depend on the experimental setup, the properties of the ground and the local meteorological data.

The model demonstrated is not applicable in non-typical days. Furthermore, it would not be applicable under rainy conditions despite the periodic trend of the variation of α-radioactivity which, nevertheless, is at a lower level than the one encountered during typical days (Seftelis et al., 2007). This is due to the fact that rain would precipitate the aerosols to the soil. A sensitivity analysis carried out on Eq. (4) has indicated that for an increase by 1% of the values of either γ-radiation or RH or T, the α-radioactivity would increase by 0.74%, 1% and decrease by 1%, respectively. Similarly, for an increase by 5%, α-radioactivity would increase by 3.6%, 5% and decrease by 5%, respectively.

The linear [$k(\mathrm{RH}/T)$] and exponential [$e^{\mu \Delta \gamma}$] terms in Eq. (6), account for the following parameters that influence the level of the α-radioactivity in air. The former accounts for the turbulent diffusion of radon in the air above the ground including the resuspension of radioisotopes in air. The latter is concerned with the radon in the ground pores, the release of radon at the ground boundary level and the deposition of radon progeny on the ground. Furthermore, these parameters are responsible for the variation of the γ-radiation originating from up to the boundary ground layer.

The variation of the measured and modelled α-radioactivity in the air is shown in Fig. 3 over the demonstrated period. Each day has been modelled separately using the measured radioactivity and meteorological data monitored within this day from 0 to 24 h. Then, the curve fitting to the experimental data yielded values for the coefficient k in Eq. (6) of 32, 29, 39, 33 and 30 for the 5 days considered. The different values of k, even for consecutive days, demonstrate the need to model independently each day rather than modelling all days using an average value of k. This is in accordance with the dependence of α-radioactivity on the meteorological conditions and γ-radiation which may differ from one day to the next. The curve fitting procedure indicated that the coefficient μ in Eq. (6) remained constant, to within 1%, at a value of 10.15 kcpm^{-1}. The average difference between the measured and modelled values of α-radioactivity, over the demonstrated period, was 1.2 Bq/m^3.

The difference between the measured and modelled diurnal variations of α-radioactivity in the air is shown in the lower part of Fig. 3. The advantage of using the variation of the difference rather than the measured one, in the interpretation of measurements related to possible nuclear emergencies, is two-fold. Firstly, the periodic form of the measured diurnal variation of α-radioactivity is now reduced to an easier to interpret almost flat response near 0. Secondly, the span between the minimum and maximum values of up to 18 Bq/m^3, encountered in the periodic form of the measured α-radioactivity, is now reduced to within 4 Bq/m^3. Then, sudden unexpected peaks of low level radioactivity would be enhanced over the natural background.

During a typical day, the α-radioactivity in air is solely due to radon released from the ground under the influence of the meteorological conditions. Then, for a measured (M_e) and modelled (M_o) values of the α-radioactivity ($M_e - M_o$) should be close to 0. The envelope defined by the error bars at (22.5% M_e) above the measured α-radioactivity is shown in Fig. 3. Any sudden unexpected peaks, deviating the difference ($M_e - M_o$) away from 0 and consequently the measured variation away from the modelled one, as well as satisfying the condition $|M_e - M_o| \geqslant (22.5\%\ M_e)$ would then be excluded as part of the natural background. These peaks could be attributed to artificial radioactivity which was accidentally or operationally released from a nuclear installation, alarming for a possible nuclear emergency. Operational releases may be due to some undeclared-clandestine-activities in an installation. In this context, monitoring of natural radioactivity in the vicinity of an installation could contribute to the early detection of such activities.

The timely detection of sudden unexpected peaks and their evaluation as a possible nuclear emergency, requires meaningful modelling on the basis of fewer measurement points soon after the appearance of the peaks. The value of coefficient k would now be based on measurements from a time period shorter than 24 h. The trend of the variation of coefficient k calculated over time periods from 0 to 4, 8, 12, 16, 20 and 24 h is shown in Fig. 4. The coefficient reaches values within 2%, 1.5%, 0.65% and 0.2% of its maximum value, which occurs after 24 h, at the end of the time

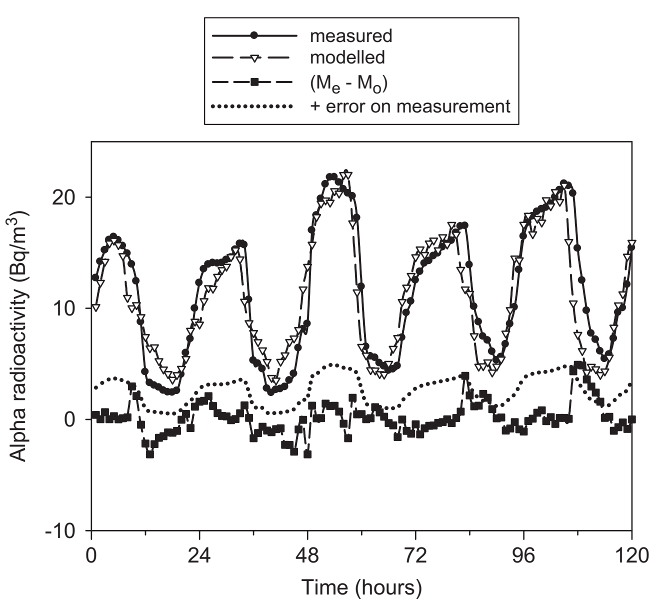

Fig. 3. Measured and modelled α-radioactivity in the air near the ground and their difference over 5 consecutive days.

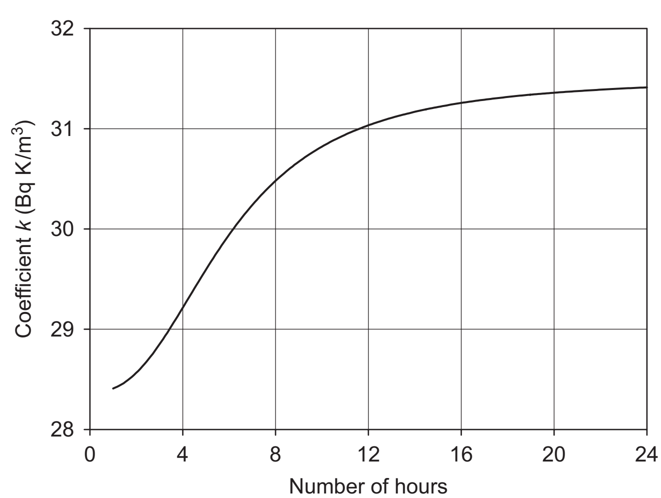

Fig. 4. Variation of the coefficient k in terms of the number of hours considered for its calculation.

periods from 0 to 10, 12, 16 and 20 h, respectively. Then, the use of these values of k from the shorter time periods, instead of its maximum after 24 h, would underestimate the modelled α-radioactivity by these amounts, on the basis of the linear dependence of the air α-radioactivity on k (Eq. (6)). Nevertheless, the periodic variation in α-radioactivity would not be affected, allowing sudden peaks above the background to be timely detected.

4. Conclusions

The diurnal variation of α-radioactivity in air and γ-radiation near the ground has been monitored. The effect of the local meteorological conditions of relative humidity and temperature, on the radioactivity levels has been investigated. The α-radioactivity in air, the γ-radiation near the ground and relative humidity reached a peak in the morning. Then, they decreased in the afternoon, followed by a rise to the peak of the following morning.

The variation of the measured α-radioactivity in air and γ-radiation near the ground, complemented by the monitoring of the local meteorological conditions, characterises this area. Hence, these measurements may be important when it is necessary to interpret sudden peaks in the radioactivity in air. These peaks may originate from the artificial release of radionuclides in the air due to a nuclear emergency. Therefore, the combined radioactivity and meteorological data monitoring, may prove valuable in the timely detection and evaluation of such an emergency.

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