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Diurnal variation of radon progeny

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Abstract

The diurnal variation of the gross alpha (α) radioactivity in the air near the ground and the gamma (γ) radioactivity emitted from the ground have been monitored in North-eastern Greece. Meteorological information comprising air temperature and humidity has been simultaneously recorded. Over a period of the 24 h of a typical day, the variation of α-radioactivity reaches a peak in the morning followed by a remarkable decrease, rising to a second peak in the afternoon. Furthermore, its significant dependence on the air temperature and humidity is confirmed, rising with an increase in humidity and decrease in temperature. The variation of the ground γ-radioactivity follows that of the air α-radioactivity. A mathematical model has been developed to describe the diurnal variation of the α-radioactivity in the air near the ground in terms of the above meteorological variables and ground level γ-radioactivity.

Keywords: Radon progeny; Alpha air radioactivity; Temperature; Humidity; Ground gamma radioactivity; Modelling

1. Introduction

Monitoring the air radioactivity is widely used as an early warning system for nuclear emergencies (Venuti et al., 1990; Ermilov and Yaryna, 1989), stemming from the fact that the fastest natural conveyor of radionuclides is air. In such an emergency, the α- and β-radioactivity due to actinides and fission products, for example from a nuclear fallout, would be detected. The level of air radioactivity in such a case would be above the natural background due to radon released into the atmosphere from the terrestrial radionuclides ^{238}U and ^{232}Th in the ground.

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Real time measurements of atmospheric radioactivity, acquired for the timely detection of nuclear emergencies, are rather challenging because of the low concentrations of the radioactivity involved. Some of the measurements are therefore accomplished through the use of an on-line air sampling via a step moving filter-tape (Smesters, 1995). These measurements can be further used to analyse the spatial and temporal variations of natural radioactivity and especially radon and its short-lived decay products. Monitoring of the concentration of radionuclides in the lower layers of air has been found to correlate with meteorological parameters such as air temperature, humidity, air speed, atmospheric electric field and airflow direction (Garcia-Talavera et al., 2001; Latha, 2003).

The work presented examines the diurnal variations of gross α-radioactivity in the air near the ground and the ground level γ-radioactivity. Furthermore, the dependence of the α-radioactivity on the γ-radioactivity and the meteorological variables of air temperature and humidity is investigated.

2. Materials and methods

2.1. Radioactivity considerations

Radon atoms from the terrestrial radionuclides ^{238}U and ^{232}Th are transported through the subsoil pore spaces and, while an amount of them will decay, the rest will be released into the atmosphere. The amount of radon depends on the amounts of ^{238}U and ^{232}Th in the ground and is influenced by the type, porosity, dampness and temperature of the soil cover. Radon (^{222}Rn) from the ^{238}U series has a long enough half-life of 3.82 days in order to diffuse out of the soil and into the atmosphere where it remains for several days. However, its decay products are typically short-lived, so they follow the distribution of the radon. The only long-lived product is ^{210}Pb with a half-life of 22.3 years. Radon and its short-lived daughters ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po are also valuable as natural tracers in the troposphere, in particular at the boundary layer near the ground (Porstendorfer et al., 1991; Sesana et al., 2006).

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. 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. While some of them are γ-ray emitters, it is expected that the ground level γ-radioactivity would in someway reflect the variation of radon release and mainly its concentration in the air near the ground.

Measurements of the diurnal variation of ground level total γ-activity have been observed to follow a periodic variation (Mojzeš, 1998). Furthermore, the diurnal variation of ^{238}U, ^{232}Th and ^{40}K were studied through γ-spectroscopy. Hence, it was found that the main contributor to the periodic variation of ground level total γ-activity is the ^{238}U progeny. The variation shows a small increase in the morning, followed by a decrease in the afternoon. The variation can be explained through the influence of the environmental conditions on the release rate and concentration of radon from the underground to the air near the ground.

2.2. Measurement of radioactivity

Measurements of radioactivity in air were carried out in the Nuclear Technology Laboratory of Democritus University of Thrace, in the city of Xanthi in North-eastern Greece (Seftelis et al., 1995).

Gross α-radioactivity concentration (Bq/m^3) was measured every 10 min on the basis of the percolation of air through a paper (tape type) filter with a pore diameter of 0.5 Å (FAG, 1989). A 2″ thick plastic type detector coated with ZnS (50 mm diameter) was positioned right on the filter in order to measure the radioactivity. The γ-radiation (cpm) from the upper ground layers was measured every 10 min, by a 3″ × 3″ NaI(Tl) detector located near the chimney at a height of 2 m above the ground. Furthermore, the air temperature (°C) and relative humidity (%) were simultaneously monitored using commercial sensors. Radioactivity measurements and meteorological data were stored every 10 min, on a personal computer via serial interfaces and an analogue to digital card for further processing.

3. Results and discussion

The variation of gross α-radioactivity in air over the 24 h of a typical day having wind speed less than 4 m/s and no precipitation is shown in Fig. 1. A peak is reached in the morning, followed by a rapid decrease to a minimum value in the afternoon. The variation rises again to a second peak of lower amplitude in the evening, followed by an increase towards the peak of the following morning. The result is consistent with other studies (Porstendorfer et al., 1991; Sesana et al., 2003; Nagarajaa et al., 2003). The minimum and maximum values encountered in the variation differ by a factor of about 3.

It has already been mentioned that some of the radon progeny emit significant γ-radiation as well as α-particles. It is therefore expected that the increase of the α-radioactivity in the air near the ground is accompanied by an increase in the ground level total γ-radiation. The variation of ground level γ-radiation over 24 h is shown in Fig. 1. The trend of the variation follows that of the α-radioactivity, with a peak in the morning and a lower value in the afternoon. In fact, the γ-radiation reflects the concentration of the radon progeny in the air near the ground rather than the rate of radon release from the soil. During a typical day, the air gross α-radioactivity should be proportional to the soil γ-radioactivity.

The diurnal periodic variation of air α-radioactivity is observed throughout the year as demonstrated from its monthly mean diurnal variation over the period of 1 year of measurements (Fig. 2). The curves are clustered in two distinct families. One family comprises the variation for the months of October, November, December and January, while the other family includes the variations of the months of March, April, May, June, July, August and September. The former family comprises the months of autumn and winter, while the latter the period of spring

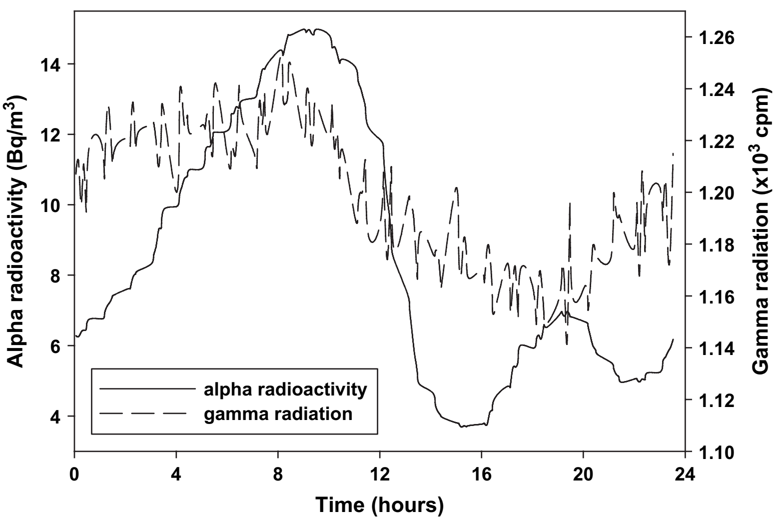

Fig. 1. Variation of air radioactivity during a typical day.

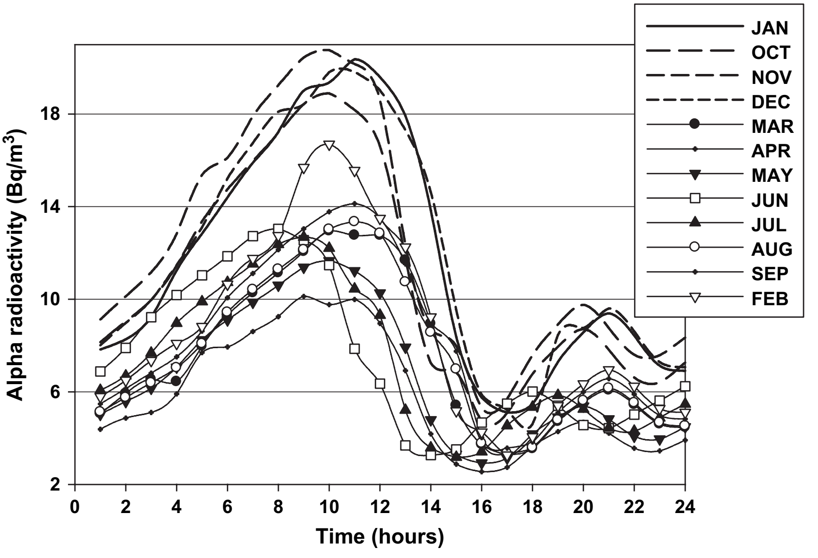

Fig. 2. Monthly mean diurnal variation of α-radioactivity throughout a year.

and summer. The curve representing the month of February falls between the two families indicating a transition from winter to spring. Furthermore, the former families of curves have a higher concentration in α-radioactivity than the latter ones, an observation that agrees with other studies (Nagarajaa et al., 2003).

The diurnal variation of radioactivity (Fig. 1) indicates a possible dependence of the radioactivity concentration in air on meteorological parameters. The temperature and relative humidity were simultaneously monitored over the 24-h period of the measurement of the α-radioactivity in air. The variations of the temperature and relative humidity over this period are shown in Fig. 3. The increase of relative humidity to a maximum in the early morning is

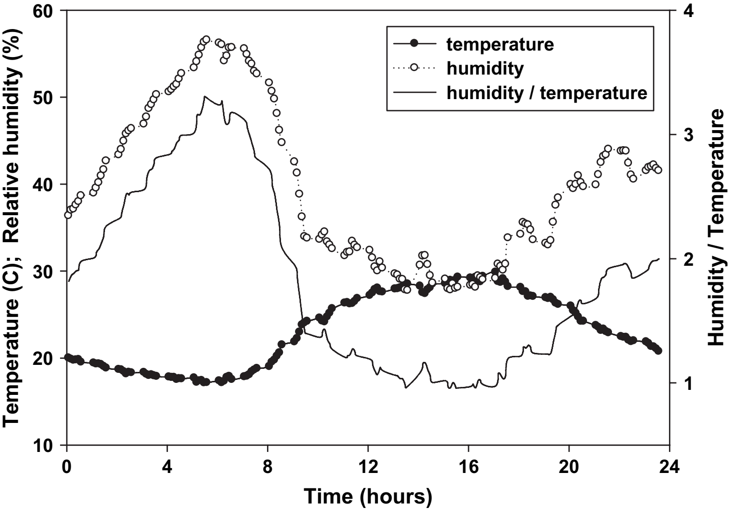

Fig. 3. Variation of temperature and humidity during a day.

followed by a notable reduction to a minimum value in the afternoon, followed by an increase towards the maximum value in the following day. The phenomenon is accompanied, as expected, by a temperature variation whose trend is the inverse of the humidity variation. Hence, the temperature decreases to a minimum value at the early morning hours and then increases to a maximum in the afternoon, followed by a decrease towards the minimum value in the following day.

The rise in temperature enhances the release of the radionuclides from the soil by drying it (Rogers and Nielson, 1991), but not the radionuclides' concentration at the layers of the atmosphere near the soil (Sesana et al., 2003). This is due to the fact that the increased airflow pushes the radionuclide particles to the upper layers of the atmosphere. Solar heating during the daytime tends to induce some turbulence, so that radon is more readily transported upwards and away from the ground. At night and in the early morning hours, atmospheric (temperature) inversion conditions are often found, which tend to trap the radon closer to the ground.

The effect of raining periods on the gross α-radioactivity in air is shown in Fig. 4 over a period of 15 days. The periodic phenomenon already demonstrated in Figs. 1 and 2 is clearly seen. However, it should be noted that in raining periods the expected diurnal radioactivity peak, although may exist as a trend, does not reach the levels achieved under typical conditions, which is in accordance with other studies (Nagarajaa et al., 2003). This is due to the fact that rain precipitates the aerosols to the soil.

The monitoring of α-radioactivity in air can be utilized as an early warning system for a nuclear emergency such as a fallout containing radioactive actinides and fission products. In such an emergency, any rainy condition would decrease, according to Fig. 4, the measured α-radioactivity from the fallout itself, besides that from the natural background. This may lead to misinterpretation of the measurement and consequently its identification as a nuclear emergency. Nevertheless, this situation will be compensated by the fact that the fission products present in the fallout would be deposited on the ground resulting in an elevated γ-radiation emission over the background, which would alarm for the nuclear emergency.

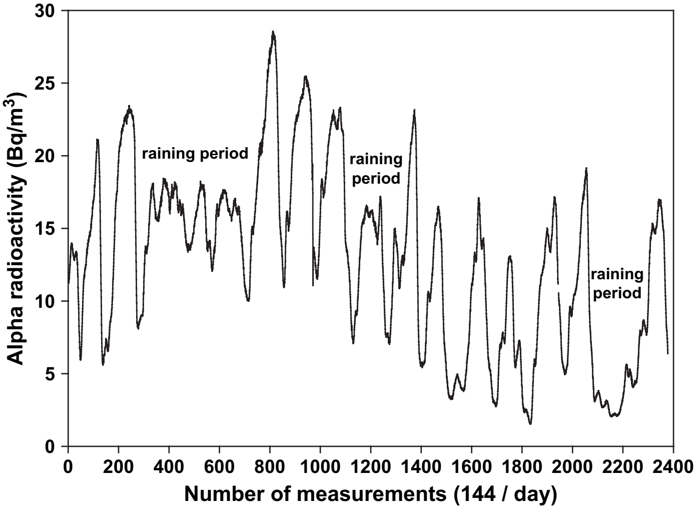

Fig. 4. Concentration of gross α-air radioactivity over 15 days.

In an ideal situation, one would like to have a flat background of α-radioactivity, above which any deviation would indicate the presence of artificial radioactivity, e.g. due to a fallout. The diurnal variation of α-radioactivity and its dependence on meteorological conditions (Figs. 1 and 3) contain minima and maxima differing by a factor of 3.5 complicating the identification over this periodic variation of any artificial radioactivity. Hence, a mathematical model was developed to describe the typical diurnal trend of the variation of the gross α-radioactivity in the air and normalise it to the desired flat background response preferably at a zero value. The model is based on the environmental variables of relative humidity and temperature, as well as the measured α- and γ-radioactivity in the air near the ground and ground level, respectively, according to the hypothesis

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

where, RH is the relative humidity (%), T is the temperature (°C), α is the α-radioactivity in the air near the ground ($\mathrm{Bq/m^3}$) and γ is the ground level γ-radioactivity (cpm).

The ratio of relative humidity to temperature is proportional to the variation of the gross α-radioactivity in air (Figs. 1 and 3). Therefore, the formulation is expected to have the form

$$\alpha = f_1(\mathrm{RH}/T, \gamma) \tag{2}$$

or,

$$\alpha = f_1(\mathrm{RH}/T) f_2(\gamma) \tag{3}$$

and hence, on the assumption that the variation is linear,

$$\alpha = \kappa(\mathrm{RH}/T) f_2(\gamma). \tag{4}$$

Furthermore, considering that the radioactive decay and hence the radioactivity concentration follow a natural exponential law, the factor f_2 is assumed to have the form

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

hence,

$$\alpha = \kappa(\mathrm{RH}/T)\mathrm{e}^{\mu\gamma} \tag{6}$$

where κ and μ are constants, which depend on the experimental setup and the ground.

The variation over 24 h of the gross α-radioactivity measured in air and predicted/normalised by the model is shown in Fig. 5. The predicted/calculated variation is mainly within an upper and a lower envelope defined by the error bars at ±22.5% around the measured values (Smesters, 1995). The average standard deviation of the experimental values is 1.33 $\mathrm{Bq/m^3}$. The variation of the difference over 24 h between the measured and predicted/calculated values of α-radioactivity is shown in Fig. 5. The periodic form of the measured α-radioactivity with a maximum variation of about 18 $\mathrm{Bq/m^3}$ is now reduced to a response of a maximum variation within 3 $\mathrm{Bq/m^3}$.

The model developed is concerned with the diurnal variation of the natural background in both air α-radioactivity and γ-radiation from the soil. The purpose of the model is to normalise the diurnal variation in a way that sudden peaks in radioactivity over the natural background are enhanced. The model has been developed for a typical day having wind speed of less than 4 m/s

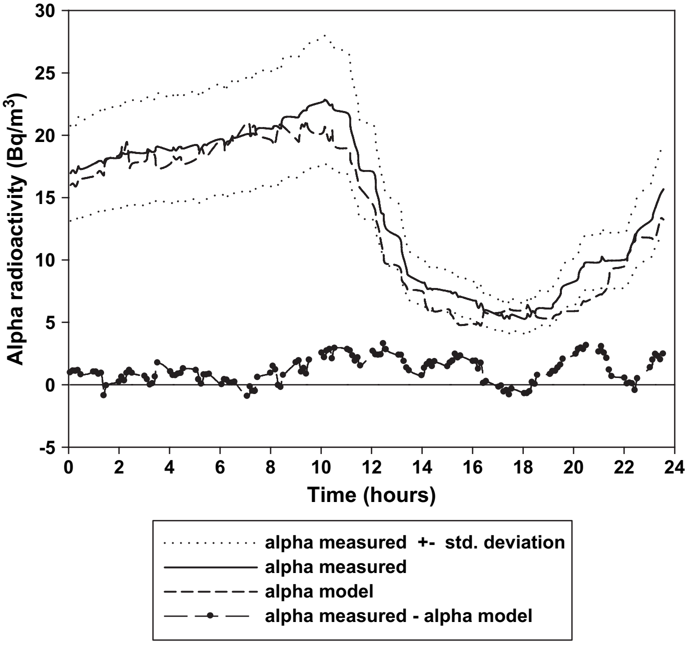

Fig. 5. Modelling of α-radioactivity in air near the ground as a function of meteorological conditions and ground level γ-radioactivity.

and no precipitation; hence, it would not be applicable in rainy conditions. Further work will be pursued to examine the influence of additional factors like air pressure and wind speed on the determination of the parameters κ and μ of the formulation used in the modelling.

4. Conclusions

The diurnal variation of air α-radioactivity and γ-radioactivity near the ground has been monitored. The effect of the meteorological conditions of relative humidity and temperature, in the area of the measurements, on radioactivity level has been investigated. The air α-radioactivity and relative humidity reached a peak almost simultaneously in the morning. The increase in temperature caused an increase of airflow from the lower (warm) to the upper (cooler) atmospheric layers, eventually resulting in the reduction of gross α-radioactivity in the air lower layers. The variation of ground level γ-radiation is proportional to the variation of α-radioactivity.

These findings may be important when one is called to interpret sudden peaks in the air radioactivity. These peaks may derive from natural reasons or from the artificial release of radionuclides in the air due to a nuclear emergency. A combined radioactivity and meteorological data monitoring may therefore prove valuable in such cases.

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