DOI: 10.1007/s10967-008-7431-0 *Journal of Radioanalytical and Nuclear Chemistry, Vol. 280, No.3 (2009) 451–455*

The diurnal monitoring of the air radioactivity using portable instrumentation

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(Received November 13, 2008)

A method is proposed which, through the monitoring and modeling of the diurnal variation of α-radioactivity in the air near the ground, appears with a potential in enhancing the measurement of low radioactivity unexpected peaks over the natural background. Portable field instrumentation was used for the monitoring which further included the total γ-radiation at ground level, the relative humidity and temperature. The variation of the α-radioactivity follows a periodic form with peaks in the morning and in the afternoon. The applicability of a mathematical model to describe this variation of the α-radioactivity in terms of the meteorological variables and the γ-radiation was tested positive. This could reduce the difference between the measured and modeled periodic variation to an almost flat one, above which sudden unexpected peaks of radioactivity from possible undeclared nuclear activities could be easier identified.

Introduction

The natural radioactivity background and its diurnal variation due to local meteorological conditions are parameters characterising a particular area. An unexpected increase in the radioactivity would cause the form of the diurnal variation of the natural radioactivity background to deviate from the one characterizing the area. The increase may originate from accidental or operational releases of radioactivity in the air or seismic phenomena.

In the case when the operational releases from a facility occur outside the designated points of a fuel cycle, they may be due to undeclared nuclear activities within the facility. Over the last decade, the scientific, technical and legislative communities have been challenged with the issue of undeclared nuclear activities in relation to non-proliferation in nuclear safeguards.[1,2] This stemmed from the need of the timely detection of such activities and their prevention from been carried out, which could eventually produce material for terrorism acts. Evidently, such activities could be accompanied by the release of radioactivity to the environment around the facility, such as the air, ground, vegetation and water. Analysis of these environmental samples could reveal the releases of radioactivity and the possible undeclared production of nuclear material.[3,4]

The air would be the initial carrier of the radioactivity, upon its release and prior to its deposition to the other environmental samples of interest. Hence, the real time monitoring of the natural background radioactivity of a particular area could be a potential way to detect possible undeclared activities, well before the deposition of the radioactivity. This stems from the fact that the unexpected increase in radioactivity would alter the form of the diurnal variation from the one of the natural background which characterizes the area, indicating possible undeclared nuclear activity. The monitoring would comprise the α-radioactivity in the air, the γ-radiation from the ground and the local meteorological conditions.

In this work, a mathematical function[5,6] is used to describe the diurnal variation of the α-radioactivity in the air in terms of the γ-radiation from the ground and the meteorological conditions in an area of interest monitored with portable field commercial instrumentation. The function could be used for the normalization of the measured variation of the α-radioactivity, in a way to enhance the measurement of sudden unexpected peaks of low level radioactivity over the natural background, which may contribute to the early detection of radioactivity releases and hence possible undeclared nuclear activities.

Experimental

The natural background, mainly due to radon (^{222}Rn) and its progeny, characterizes a particular area in terms of the concentration of ^{238}U and ^{232}Th in the ground and its geological characteristics.[7] Therefore, any unexpected increase in the radioactivity above this background could be attributed to some release of artificial radionuclides in the air, possibly stemming from some undeclared activity.

Radon 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. Then, the α-radioactivity in the air near the ground would be due to the short-lived progeny of ^{222}Rn, namely ^{218}Po, ^{214}Pb and ^{214}Bi. Among the daughters of ^{222}Rn, ^{214}Pb and ^{214}Bi are significant γ-radiation emitters with energies ranging between 0.242 MeV and 2.204 MeV. Then, monitoring of the natural background would include measurements of the α-radioactivity in the air and the γ-radiation from the ground.

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0236–5731/USD 20.00

Akadémiai Kiadó, Budapest
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Simultaneous measurements of the α-radioactivity in the air near the ground and the total γ-radiation at ground level were carried out by the Nuclear Technology Laboratory of Democritus University of Thrace, in the city of Xanthi in North-eastern Greece. Furthermore, the relative humidity and temperature of the air were measured simultaneously, since they are parameters on which the radioactivity is heavily dependent.[7] Portable field instrumentation was used which was logged on a notebook for data collection and their subsequent evaluation.

The α-radioactivity ($Bq \cdot m^{-3}$) was measured using a portable radon monitor comprising the Pylon AB-5 fitted with a Lucas A300 cell. The AB-5 is a lightweight radiation monitor with a built-in photomultiplier tube, air pump and a rechargeable battery to in-situ remote measurements. The cell is a gas chamber, with a volume of 300 cm^3, which is coated with zinc sulphide activated with silver (ZnS(Ag)) acting as a scintillation detector. The air sample is pre-filtered before reaching the chamber where the α-particles emitted during the decay of the radon progeny are measured. The continuous mode was employed with AB-5, where air was flown through the cell continuously. Measurements of a 10 minute duration were taken at 10-minute intervals, 3.5 hours after the start of the air flow through the cell and for a period of 24 hours.

The total γ-radiation at ground level was measured with the portable Thermo-scientific IdentiFINDER spectrometer. The instrument comprises a 35 mm diameter by 51 mm long NaI (Tl) detector. It integrates a high voltage power supply, an amplifier and multichannel analyzer for the gamma-ray spectroscopy. Furthermore, it incorporates a GM tube for high gamma dose rate measurements. The γ-radiation ($kc \cdot h^{-1}$) from the ground layers was measured every 10 minutes, through evaluation of the gross area of the gamma-ray spectrum in the energy region 40 keV to 2.5 MeV. The air temperature (°C) and relative humidity (%) were simultaneously monitored using commercial sensors. Measurements were stored on a personal computer for further processing.

Results and discussion

The simultaneous measurements of the α-radioactivity, γ-radiation and environmental parameters were carried out over a period of several consecutive typical days having a wind speed less than 4 $m \cdot s^{-1}$ and no precipitation. The measurements confirmed the characteristic of such a day, namely the periodic variation of the diurnal radioactivity rising in the morning and decreasing in the afternoon. The periodic form encountered in the variation of α-radioactivity is consistent with other studies.[5,9,10] The diurnal variation of the α-radioactivity for one of the days of the measurements is shown in Fig. 1. Two peaks are observed in this periodic variation, one in the morning, followed by a rapid decrease to a minimum value in the afternoon and a second peak of lower amplitude in the evening. Then, the variation rises further towards the peak of the following morning.

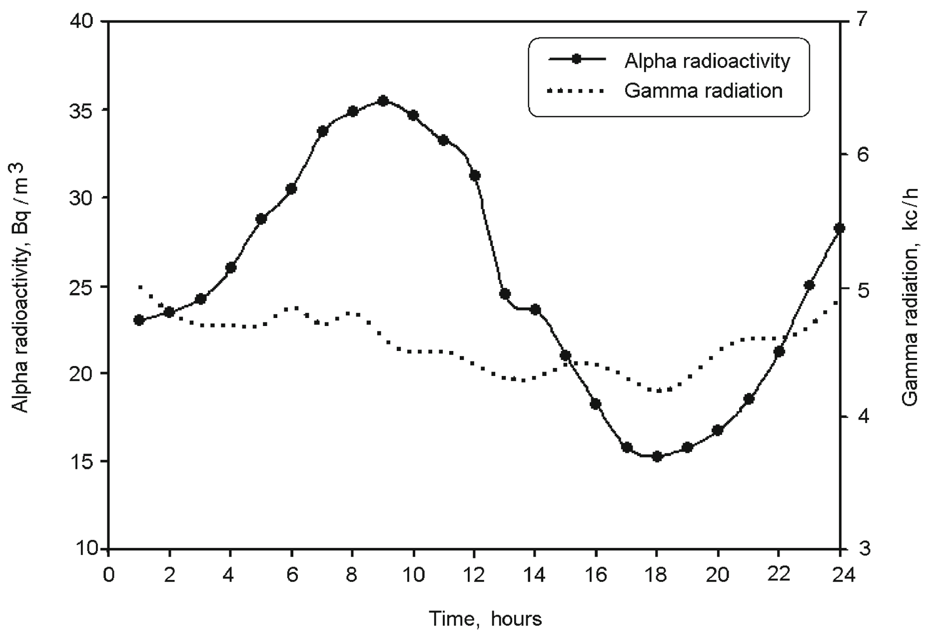

Fig. 1. Diurnal variation of environmental radioactivity and meteorological conditions

It has been mentioned that some of the radon progeny are significant emitters of α-particles and γ-radiation. Hence, any increase of the α-radioactivity in the air near the ground would be accompanied by an increase in the γ-radiation from the ground. The diurnal variation of the γ-radiation at ground level is shown in Fig. 1. The trend of the variation follows that of the α-radioactivity, having a peak in the morning decreasing to a lower value in the afternoon and then rising towards the peak of the following morning.

The diurnal variation of radioactivity (Fig. 1) indicates a possible dependence of the radioactivity concentration in air on meteorological conditions. The temperature (T) and relative humidity (RH) were simultaneously monitored over the same 24-hour period. The diurnal variations of the temperature and relative humidity, measured simultaneously with the α-radioactivity and γ-radiation, and their ratio over this period are shown in Fig. 2. The relative humidity reaches a maximum in the early morning and then decreases to a minimum value in the afternoon, followed by an increase towards a maximum value in the following day. The trend of the temperature variation is the inverse of the relative humidity variation.

The meteorological variations precede those of the α-radioactivity and γ-radiation, which is in accordance with the fact that temperature and humidity are responsible for the movement of radionuclides in the ground and their release to the air. Hence, according to the data analysis over the demonstrated 24-hour period, the α-radioactivity and γ-radiation variations lag by 1–2 hours the variations of (RH/T).

The increase in temperature dries the soil, enhancing the release of the radionuclides from it. However, it does not enhance the concentration of the radionuclides at the layers of the atmosphere near the soil.[10] This is because radionuclide particles are pushed to the upper layers of the atmosphere due to the increased air flow. During daytime, solar heating tends to induce some turbulence causing the radon to be more readily transported upwards and away from the ground. At night and in the early morning hours, atmospheric temperature inversion often occurs causing trapping of the radon closer to the ground.

The periodic form of the variation of α-radioactivity contains minima and maxima differing by a factor of 2.3, an issue complicating the identification of any low-level unexpected artificial radioactivity over this periodic variation. Ideally, one would like to have a flat background of radioactivity close to zero, above which deviations would indicate the presence of artificial radioactivity, e.g. from undeclared nuclear activities. The flat background would render easier to interpret deviations, while been close to zero would improve the minimum detection limit of the measurements.

Hence, the applicability of the mathematical model developed by SEFTELIS et al.,[5,6] which described the diurnal variation of the α-radioactivity monitored by laboratory instrumentation, was tested in this work where portable field instrumentation is employed.

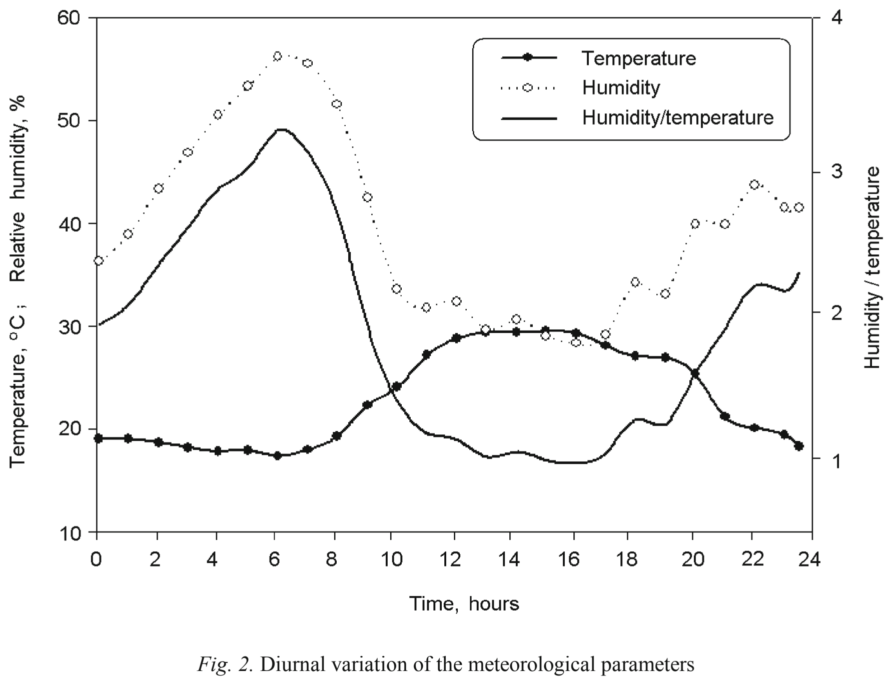

Fig. 2. Diurnal variation of the meteorological parameters

The model, aiming to reduce the variation of the α-radioactivity to the desired flat background response, is based on the fact that the daily variation of the α-radioactivity and γ-radiation depend on the relative humidity, temperature in the ground level and is described by 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 (Bq·m^{-3}) and γ is the ground level γ-radiation (kc·h^{-1}). Considering that the α-radioactivity in air ratio is proportional to the ratio of relative humidity and temperature, and follows the natural exponential law of decay, Eq. (1) becomes

$$\alpha = k \cdot (\mathrm{RH}/T) \cdot e^{w \cdot \gamma} \tag{2}$$

where k and w are constants which depend on the experimental setup and the ground.

The diurnal variations of the measured and predicted by the model α-radioactivity, as well as the difference between them, are shown in Fig. 3 over a period of three consecutive typical days. Each day was modeled separately using the measured γ-radiation and the necessary meteorological parameters monitored over the 24 hours of this day. Firstly, the periodic form of the measured α-radioactivity is now reduced to an easier to interpret almost flat response near zero. Secondly, the periodic form of the measured α-radioactivity with a maximum variation of about 18 Bq·m^{-3} is now reduced to the easier to interpret close to a flat response of a maximum variation within ±5 Bq·m^{-3}. The average difference between the measured and modeled values of α-radioactivity, over the demonstrated period of three days, is 1.5 Bq·m^{-3}.

The model has been developed for a typical day having wind speed of less than 4 m/s and no precipitation; hence, it would not be applicable in rainy conditions. Further work would be pursued in order to examine the influence of further factors such as air pressure and wind speed on the determination of the parameters k and w of the formulation used in the modeling.

Conclusions

The diurnal variations of the α-radioactivity in the air near the ground, the total γ-radiation at ground level, the relative humidity and temperature over consecutive typical days have been simultaneously monitored using portable field instrumentation. The variation of the radioactivity follows the meteorological parameters, reaching a peak values in the morning and the afternoon.

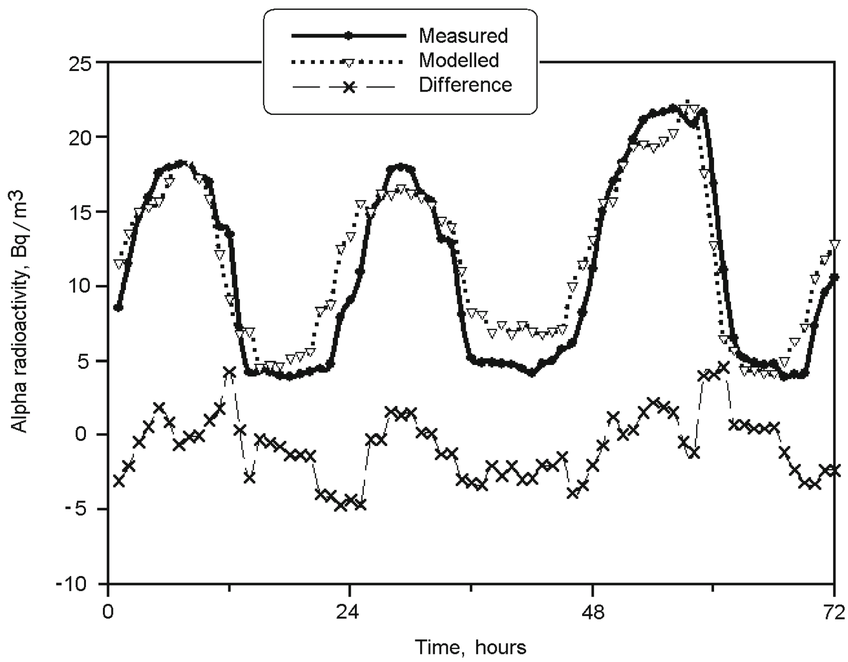

Fig. 3. Modeling of α-radioactivity in air near the ground as a function of the meteorological parameters and ground level γ-radioactivity over the demonstrated period of typical days

The modeling of the variation of the α-radioactivity was carried out on the basis of the other monitored parameters. The variation of the difference between the measured and modeled α-radioactivity has resulted to an almost flat variation, easier to interpret over the complicated periodic variation of the measured α-radioactivity. Furthermore, an improved minimum detection limit for the measurement of sudden unexpected peaks of low radioactivity has been obtained. Hence, the combined measurement and modeling of the environmental radioactivity appears a potential method in the interpretation of unexpected increases in the air radioactivity which may have been derived from the release of artificial radionuclides in the air due to undeclared nuclear activities.

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