On the natural α-activity of the air near the ground

Part A. *Method of Measuring and a few Preliminary Results*

By S. ISRAELSSON, E. KNUDSEN, E. UNGETHÜM and L. DAHLGREN, *Institute of Meteorology, University of Uppsala, Sweden*

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ABSTRACT

A method has been developed by which the concentration of the natural α-activity of the air near the ground can be studied over a wide range of variation. The method is based on an aspiration process. The apparatus is mainly used in order to measure rapid fluctuations of the α-activity of the air near the ground. The preliminary results presented in this paper show a connection between the fluctuations of the natural α-activity and the temperature probably due to a connecting link—the vertical motion. Further simultaneous measurements of the natural α-activity and the temperature in the layer up to 3 metres above the ground have been made. The gradients were fairly steep near the ground and decreased rapidly with height.

Introduction

The study at Marsta Observatory of atmospheric radioactivity and its influence upon atmospheric electricity was, up till 1970, concerned with variations having periods greater than 1 hour. In order to study more rapid oscillations of natural radioactivity in the atmosphere near the ground, an apparatus has been constructed, which permits the study of frequencies as high as 1 cycle per 30 seconds.

In the present paper we give a description of the method of measuring rapid fluctuations of natural α-activity in the air near the ground. We also give some theoretical considerations of the oscillations of natural radioactivity in the air. A few preliminary results from measurements made in the summer of 1970 are also presented.

Theory of the variation of natural radioactivity of the air in the surface layer

Radon and thoron are formed in the ground. By exhalation from the pores in the surface layers of the ground they are set free in the atmosphere, where they bring about a series of radioactive daughter products which rapidly adhere to the aerosols of the air.

The mathematical treatment of the variation with time of the natural radioactive gases in the air is analagous with the treatment of turbulent fluctuations, i.e. we write

$$C = \bar{C} + C' \tag{1}$$

where

C = concentration of the natural radioactive gas,
$\bar{C}$ = mean or weighted mean,
C' = fluctuations from the mean value.

Similarly we obtain for wind velocity and air density

$$V = \bar{V} + V'$$

$$\varrho = \bar{\varrho} + \varrho' \tag{2}$$

where (′) denotes the deviation from the mean value (⁻).

According to the theory of diffusion of radioactive gases (see e.g. Jacobi et al., 1959, Bolin, 1962; Fontan et al., 1966), we obtain the variation with time t of the concentration C, i.e.

$$\frac{dC}{dt} = P(r, t) - \lambda C \tag{3}$$

where

$P(r, t)$ = production and/or removal of radioactive gas at the point r.
λ = radioactive decay constant

With the aid of

$$\frac{dC}{dt} = \frac{\partial C}{\partial t} + V\nabla C$$

and the equation of continuity

$$\frac{\partial \varrho}{\partial t} + \nabla(\varrho V) = 0$$

With Eqs. (1) and (2) Eq. (3) can be written

$$\frac{\partial C}{\partial t} + V\nabla C + \frac{1}{\bar{\varrho}}\nabla(\overline{\varrho C' V'}) - P(r, t) + \lambda C = 0 \quad (4)$$

supposing

$$\frac{\partial \bar{\varrho}}{\partial t} + \nabla(\bar{\varrho} V) = 0 \text{ and } P = \bar{P}$$

From the theory of turbulence we may now introduce eddy exchange coefficients. We will only consider variations of C and V in the z-direction supposing no horizontal gradients. The expression $\overline{\varrho\, C' V'}$ in Eq. (4) can then be written

$$= \overline{\varrho C' w'} = \bar{\varrho} K_z \frac{\partial C}{\partial z} \quad (5)$$

where

w' = fluctuations of the vertical velocity w
K_z = eddy exchange coefficient for variations in the z-direction

The left part of Eq. (5) has been applied in connection with gradient measurements of Rn and Tn (see Israelsson et al., 1972).

The statistical expression for the spatial structure of the variation of natural radioactivity can be given by a space correlation between the concentrations at two points a specified distance apart. The problem is almost identical with the analogous study of velocity and temperature (see e.g. Taylor, 1935).

If the fluctuations of the radioactive gas C', are measured simultaneously at two points the ocrrelation coefficient $r(x)$ is defined by

$$r(x) = \frac{\overline{C'(x_1) C'(x_2)}}{\overline{C'^2}} \quad (6)$$

$r(x)$ should generally decrease with increase of the distance X.

A time-correlation coefficient, $r(t)$, usually referred to as an auto-correlation coefficient, may also be defined in terms of the fluctuations of the radioactivity at a fixed point but separated in time by the value t (see e.g. Panofsky & Brier, 1958).

$$r(t) = \frac{\overline{C'(t_1) C'(t_2)}}{\overline{C'^2}} \quad (7)$$

It is of great value to compare the fluctuations of the natural radioactivity with meteorological parameters, e.g. temperature.

From continuous records of natural radioactivity and temperature the time-correlation function (cross-correlation function) can be computed. It is expressed by

$$r_{cT}(L) = \frac{\overline{C'(t) T'(t + L)}}{\overline{C'^2(t) T'^2(t)}} \quad (8)$$

where L = time-lag.

It is also of interest to compare corresponding expressions for radioactivity and atmospheric-electrical parameters.

Method of measuring natural α-activity in the air

(a) *Principle of the measurement of α-activity*

The method used by us is in principle according to the method described by H. Israël & G. W. Israël (1966). Atmospheric air, after having been cleansed of aerosol particles, radioactive decay products, and small ions, is aspirated through a cylindrical ionization chamber with a volume of 24 liters. The electric current created in the chamber is due to the ionization caused by the natural α-activity of the air and by the background radiation. The α-activity arises from the disintegration of radon, thoron and their decay products. Keeping out both radon and thoron, the background radiation can be measured. It is then possible to determine fluctuations of the α-activity within different frequency ranges.

(b) *Description of the apparatus*

Fig. 1 shows the arrangement of the apparatus for measuring the natural α-activity. Atmos-

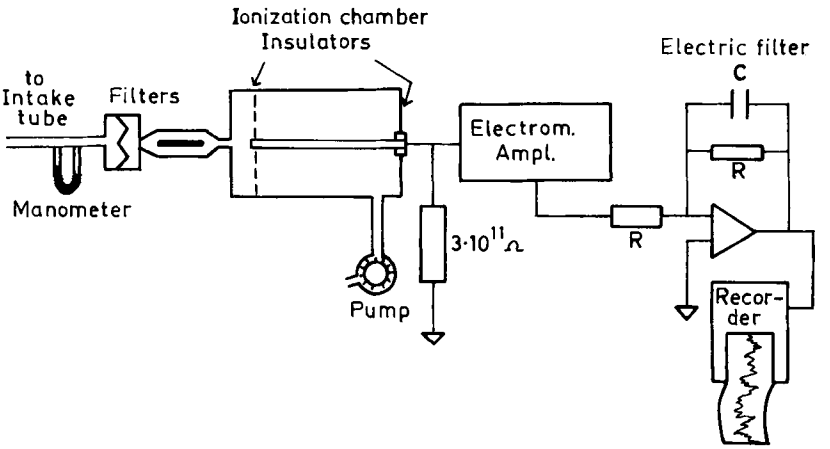

Fig. 1. Schematic arrangement of the apparatus.

pheric air is forced through an aerosol filter and through a cylinder condenser attached to a voltage source. In this way the air can be cleansed of aerosol particles, decay products, and small ions. The volume of the cylindrical chamber is only 24 liters. The outer electrode is attached to a variable voltage source and the inner electrode is earthed over a high-meg-resistance, 3×10^{11} ohm, and connected to an electrometer amplifier. The limit of the electrometer is about 10^{-13} A with an accuracy of ± 1 %. Different condensers can be connected in order to give different smoothing of the recording. The time constants are 5, 10, 25, 40, 60, 100 and 300 sec.

A detailed description of the apparatus will be given in a report, Israrelsson et al. (1972).

(c) *Effects of variable air flow through the chamber upon the ionization current*

In order to record rapid fluctuations of the α-activity the ionization chamber must be small in size. But the sensitivity of the equipment is a function of the volume. Supposing a thoron concentration of N atoms per volume unit of air and a residence time t_r of the air in the chamber, there will occur n disintegrates of thoron atoms per second given by the following equation (H. Israël & G. W. Israël, 1966).

$$n = \frac{NV}{t_r}[1 - \exp(-\lambda_{Tn} t_r)] \qquad (9)$$

where

V = volume of the chamber
λ_{Tn} = decay constant of thoron

The number of disintegrated atoms per second, n, is thus nearly proportional to V and $1/t_r$.

The volume V was in our case 24 liters, t_r can be determined for a special air flow.

There is a loss of α-activity on the way from the intake point to the ionization chamber. It can be computed, knowing the disintegration constant, the rate of air flow and the distance between the intake point and the ionization chamber. Suppose the concentrations of radon and thoron at the intake point are C^1_{Rn} and C^1_{Tn} respectively. The concentration at the entrance to the ionization chamber is thus a function of wind speed in the intake tube given by

$$C_{RT}(u) = C^1_{Rn} \exp\left[-\lambda_{Rn}\frac{l}{u}\right]$$

$$C_{Tn}(u) = C^1_{Tn} \exp\left[-\lambda_{TR}\frac{l}{u}\right] \qquad (2)$$

where

u = air speed in the intake tube
λ_{Rn} and λ_{Tn} = disintegration constants of radon and thoron respectively
l = length of the intake tube

Increasing air speed u results in an increase of C_{Rn} (u) and $C_{Tn}(u)$. All u-values gives approximately constant value C^1_{Rn} of C_{Rn} (u) due to the long half-life of radon (3.8 days). C_{Tn} (u), however, increases rapidly with u due

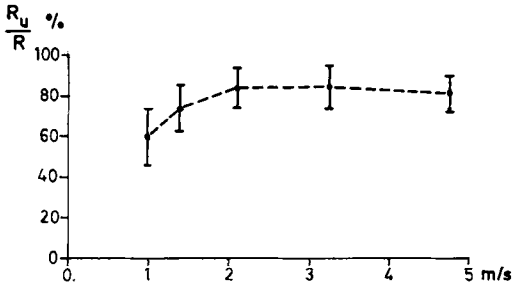

Fig. 2. The ionization current R_u as a function of the air velocity u in the intake tube. The ionization current obtained from rapid filling of the ionization chamber is R.

to the short half-life (54.5 sec). The total recorded α-activity concentration ought to increase with increasing u-values.

Due to the fact that the ionization chamber is an aspiration cylinder condenser there must be ions which do not reach the inner electrode, i.e. we measure an ionization current which is too small.

In the present investigation we have studied the effect of variable air speed in the intake tube on the ionization current. We have calculated the experimental differences between the values obtained from the usual method of recording α-activity in the air (see Israelsson et al., 1972) and from the method presented in this paper. Fig. 2 shows the ratio between the ionization current R_u obtained from the experiments with variable air velocities u and the ionization, R, obtained immediately after the ionization chamber has been filled rapidly. The quotient R_u/R is plotted against the wind speed in the intake tube. The data refer to 12 measurements.

The range bars in the figure represent the standard deviations. With wind speed u less than 2–3 m/s the ratio R_u/R increases with u; with higher values of u the ratio remains nearly constant.

In order to obtain the concentration of α-activity per volume of air from the recorded α-activity the observation series begin and finish with measurements of the concentration of radon and thoron in the intake point.

(d) *Calibration and calculation of α-concentration in ionization chamber*

The ionization chamber was calibrated with the help of radon (Rn^{222}). The activity of radon was measured at Statens Strålskyddsinstitut, Stockholm 60, with an accuracy of ±6 %. In the calibrations the ionization current was determined versus the radon content. From the calibrations the detection limit was found to be approximately 0.1 pCi/l.

Due to the fact that there is a loss of α-activity in the intake tubes and the chamber was used as an aspirated condenser we apply corrections for the influence of the rate of air flow through the chamber upon the measured ionization current given in Section (e). The systematic error affecting the measured α-activity per volume of the air should not exceed ±15 % over much of the variation range. For small α-activity concentrations the error introduced by the current fluctuations increases.

(e) *The time constant of the measuring equipment*

Determinations of the fluctuations of the α-activity in the atmospheric air require a knowledge of the time constant of the *total* measuring system. According to Fig. 1 it is possible to connect different filter condensers to measuring apparatus. These filters will lead to different smoothing of the recording.

To obtain an idea of the size of the time constant of our measuring equipment we rapidly changed the concentration of the α-activity in the air-intake point and studied after that the ionization current on the recorder. The rapid change of the α-activity in the air-intake point was obtained by placing the air-intake 2 m above the ground and after a while rapidly move it to the ground surface. Measurements of the α-activity of the air in surface layer show a remarkable vertical gradient. This is due to the increase with height of the eddy-exchange coefficient in combination with the radioactive disintegration. From the recorded ionization current we thus can determine the time constant of the whole equipment. By varying the air flow u we can determine the time constant as a function of u. For wind speeds near 5 m/s in the air intake tube we obtained from 16 measurements an average time-constant of the equipment of 6 sec. The standard deviation of the individual observations was approximately 1 sec.

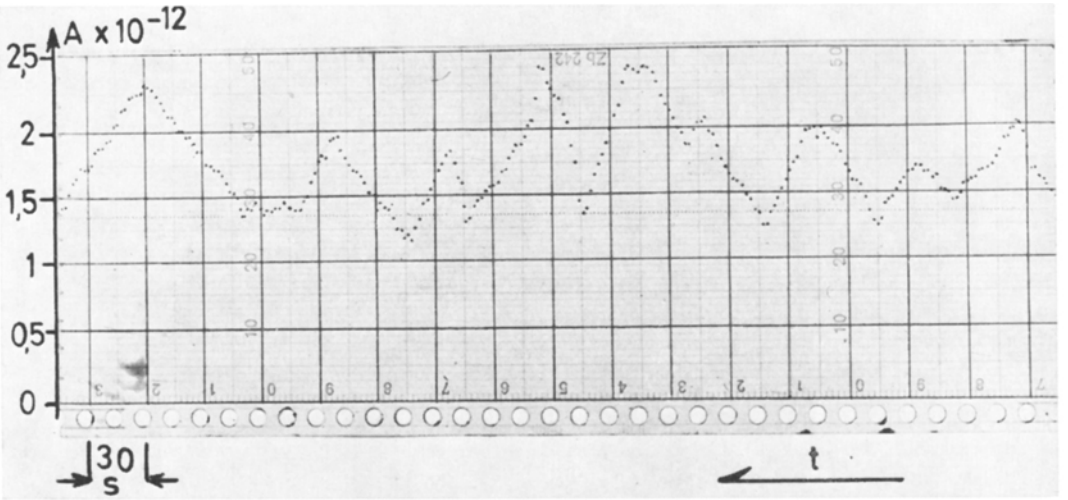

Fig. 3. Record of ionization current; α-activity and background radiation involved.

(f) *Possibilities of measuring rapid fluctuations of α-activity in surface layer*

The fluctuations of the background radiation are of fundamental importance in the study of fluctuations of the α-activity in the air near the ground. The radioactivity of the air in the surface layer is high in the Uppsala region. The background radiation is therefore low in comparison with the air activity. Furthermore, the background radiation is relatively constant.

Comparisons between fluctuations of the ionization current due to background radiation and air radioactivity show that the former radiation source in fact gives a relatively small contribution to the total ionization current. Fig. 3 shows a recording of the variation of the ionization current due to the α-activity and background radiation. The electrical time constant is here 5 sec. The intake tube was placed 1 m above the ground. Fig. 4 refers to measurements of background radiation

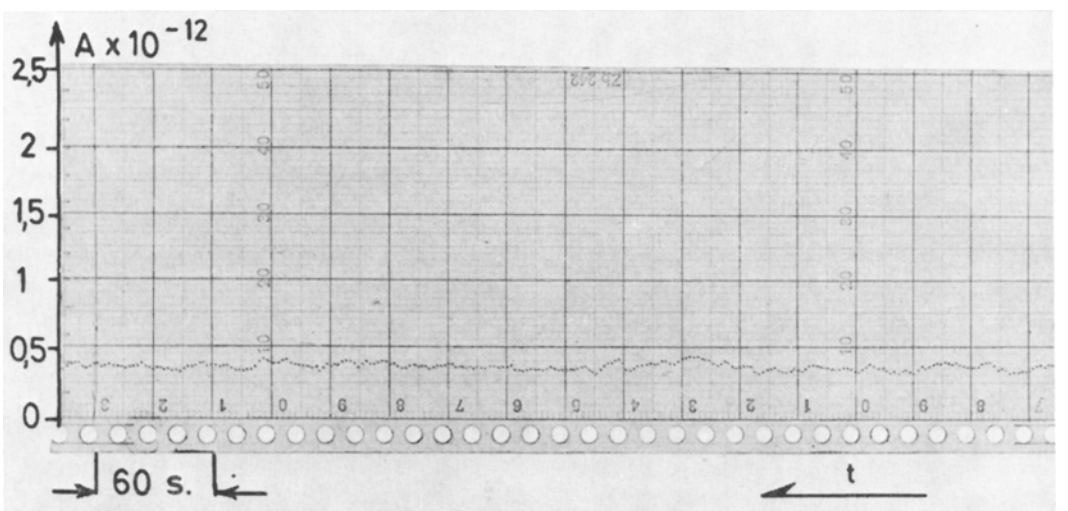

Fig. 4. Record of ionization current; only background radiation involved. Radon, thoron and their decay products are kept out of the chamber.

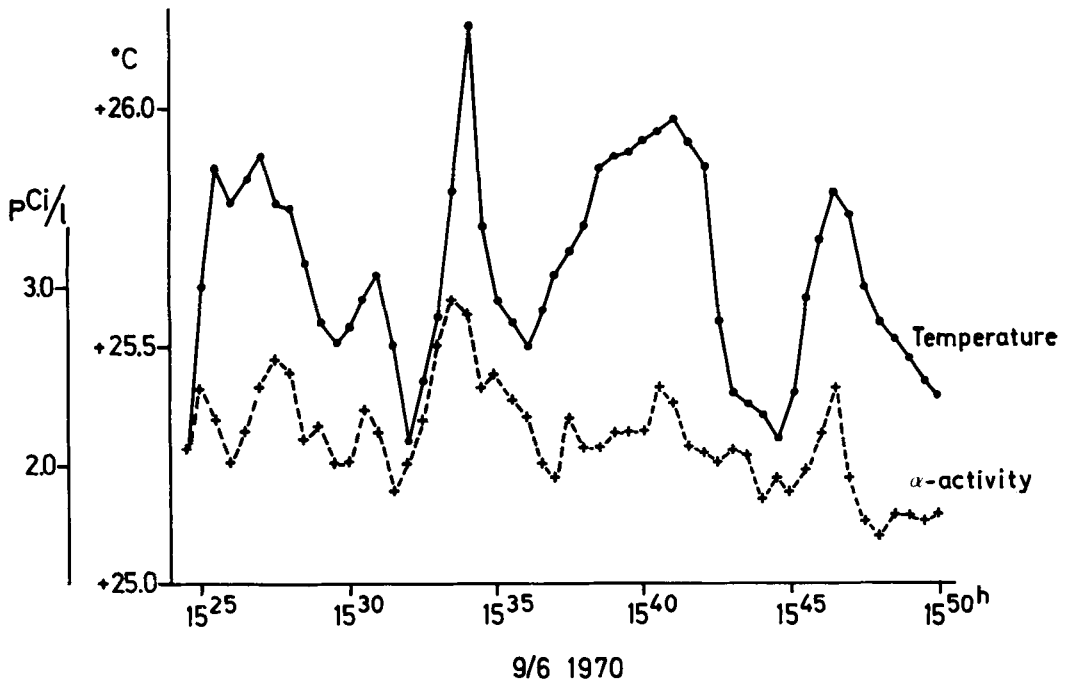

Fig. 5. Simultaneous fluctuations of natural α-activity and temperature on a clear day in summer.

alone; radon, thoron and their decay products have been kept out of the chamber. The diagrams show that the oscillations of ionization current due to background radiation have very obviously less amplitudes in the fluctuations than in the case with air activity plus background activity.

Measurements have shown that the fluctuations caused by the background activity seem to be less than 10 % of the fluctuations of the α-activity per volume of air. The influence of the background radiation are of minor or no importance when the measurements refer to frequencies below 1 period per hour. Frequencies higher than 1 period per 30 sec have not been studied. The time constant of the equipment and the statistical fluctuations of the ionization processes are too great for that purpose.

Simultaneous measurements of natural α-activity and air temperature in surface layer

From the theory of atmospheric turbulence we know that turbulence near the ground varies from one situation to another not only in intensity but also in character. The meteorological conditions affect the natural α-activity in the surface layer. Variations of natural β-activity in the frequency range below 1 period per hour have previously been studied at the Marsta Observatory and the connection with meteorological parameters has been shown (Hedenmalm, 1963; Israelsson, 1968 and 1970). For technical reasons it has not been possible to study variations of the β-activity in the frequency range above 1 period per hour whereas with the method described in the present paper fairly rapid fluctuations in the natural α-activity can be recorded.

In the present investigation we have studied the connection between the fluctuations of the natural α-activity and the temperature 1 m above the ground.

The temperature was measured with an accuracy of ±0.1°C with a ventilated copper vs. constantan thermocouple attached to the intake-tube of the ionization chamber. The thermocouple was coloured white. The α-α-activity concentration and the temperature were determined every 30 sec.

Figs. 5 and 6 show two segments of records

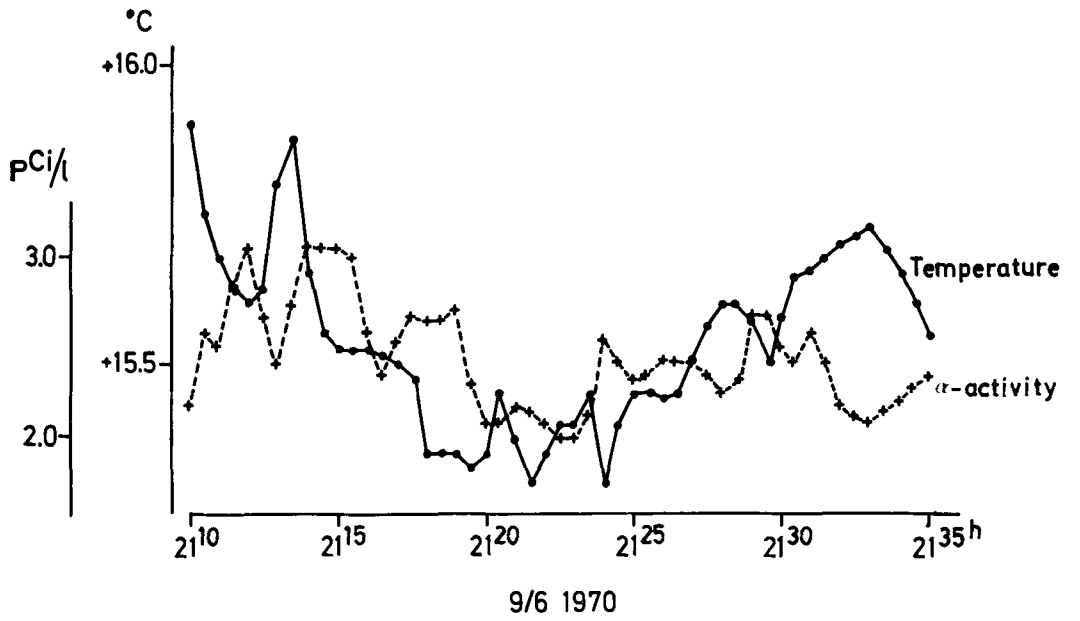

Fig. 6. Simultaneous fluctuations of natural α-activity and temperature on a clear night.

of simultaneous fluctuations of natural α-activity and temperature obtained on a clear summer day (Fig. 5) and on the following clear night (Fig. 6). The fairly high degree of correlation between the two elements must be due to the dominance of some connecting link—apparently the vertical motion.

In Fig. 5 a high temperature often occurs simultaneously with a high natural α-activity. Because of the insolation we obtain a great lapse-rate near the ground. Vertical motions, viz. transport of heated air from the ground surface upwards, causes a temperature rise in the measurement level. The air originating from the ground surface brings a high concentration of radioactive gases exhaled from the ground. Under surface inversion conditions (Fig. 6) any vertical motion upwards ought to give a temperature fall in the measurement level, but we still ought to obtain an increase in α-activity. With motion downwards the temperature rises but the concentration of the α-activity decreases.

A quantitative expression of the fluctuations of temperature and natural α-activity is provided by cross-correlation of the two variables.

From Eq. 7 we can determine the auto-correlation of natural α-activity. Fig. 7 shows some auto-correlation correlogram from 9–10 June, 1970. The period represents fine weather conditions.

The diagram shows auto-correlograms for time-lags up to 7 min. The curves (*a*) and (*b*) represent unstable stratification (daytime) from 9th and 10th June respectively. (*c*) and (*d*) represent surface inversion conditions from the same days.

In the terms of earlier discussion of the correlogram the initial decay of the correlation implies a scale of fluctuations corresponding to 2 to 4 min.

The situations with unstable stratification,

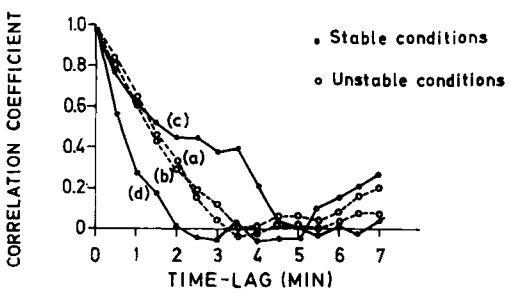

Fig. 7. Auto-correlations of natural α-activity at a height of 1 m (9–10 June, 1970). Sampling-period 45 min. Each autocorrelogram represents 2 sampling periods.

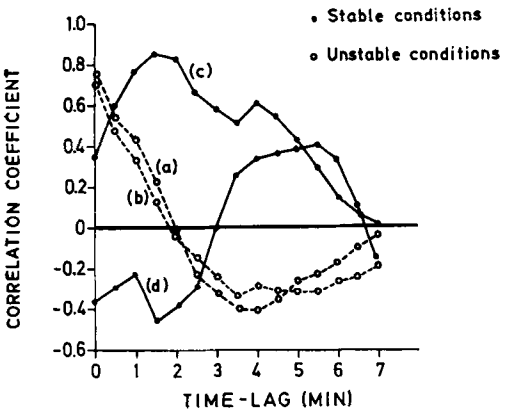

Fig. 8. Cross-correlations of natural α-activity and temperature at a height of 1 m. The data refer to the same period as Fig. 7.

curves (*a*) and (*b*) show comparable correlograms with a scale of fluctuation corresponding to about 2 min. The situations with stable stratification, however, show a more rapid initial decay corresponding to a smaller scale of fluctuation. The two curves diverge, however, For time-lag 2–4 min the auto-correlation is about +0.4 for curve (*c*) and 0.0 for curve (*d*).

In Fig. 8 we have determined the cross-correlation of natural α-activity and temperature according to Eq. 11. The calculations include time-lags up to 7 min. The data refer to the same period of observation as Fig. 7. Curves (*a*) and (*b*) represent situations with unstable stratification, (*c*) and (*d*) situations with stable stratification.

The correlograms representing unstable and stable stratification do not coincide. Unstable conditions show an initial decay. Time-lag of 0 min gives a correlation coefficient +0.7 to +0.8. The cross-correlation function shows the lowest values for time-lags 4–5 min, viz. −0.3 to −0.4. With longer time lags the cross-correlation coefficient approaches the value zero.

The two situations with stable stratification, however, show initially an increase in correlation. The individual curves differ remarkably. Curve (*c*) shows a maximum value (+0.85) for time-lags of 1.5 min and a less obvious peak at time-lags of 4 min. Curve (*d*) has a pronounced maximum value, +0.4, for time-lags between 4 and 6 min and a less pronounced peak-value for the time-lag of 1 min.

The correlograms show the difference in the variations of the natural α-activity and the temperature, which was discussed above. With unstable stratification the curves of temperature and α-activity seem to coincide fairly well, but with stable stratification the two parameters seem to have an opposite variation, i.e. maximum of temperature concides with minimum of α-activity, and the inverse.

As a conclusion we may state that there thus seems to be a connection between rapid fluctuations of natural α-activity and temperature.

In the near future we will study more closely the connection between the fluctuations of natural radioactivity and other usual meteorological parameters for frequencies higher than 1 period per 30 min, according to the theoretical consideration given in the present paper.

Variations of natural α-activity and of temperature in 0–3 m layer

The intensity of the atmospheric turbulence depends to a high degree upon the vertical gradient of temperature. In the previous section we have studied the connection between the fluctuations of temperature and natural α-activity in the air near the ground. The variation of the temperature and the natural α-activity within the 0–3 m layer is the subject of the following study.

The diurnal variation of temperature in the lowest layers of the atmosphere is often pronounced in clear weather situations. During the hours of daylight from shortly after dawn to about 1 hour before sunset temperature generally decreases with height, fairly rapidly in the lowest layers, more slowly in higher layers. At night, stability is greater and very often the temperature increases with height, the gradient again being steepest in the lowest layers.

In overcast conditions or in situations with strong winds, the diurnal variation of the temperature profile almost entirely disappears.

There is an interrelationship between temperature profile and wind profile owing to equality between eddy viscosity and eddy conductivity. The connection between temperature and wind profiles under conditions of free convection and neutral stability has been postulated by Monin & Obukhov (1954). According to

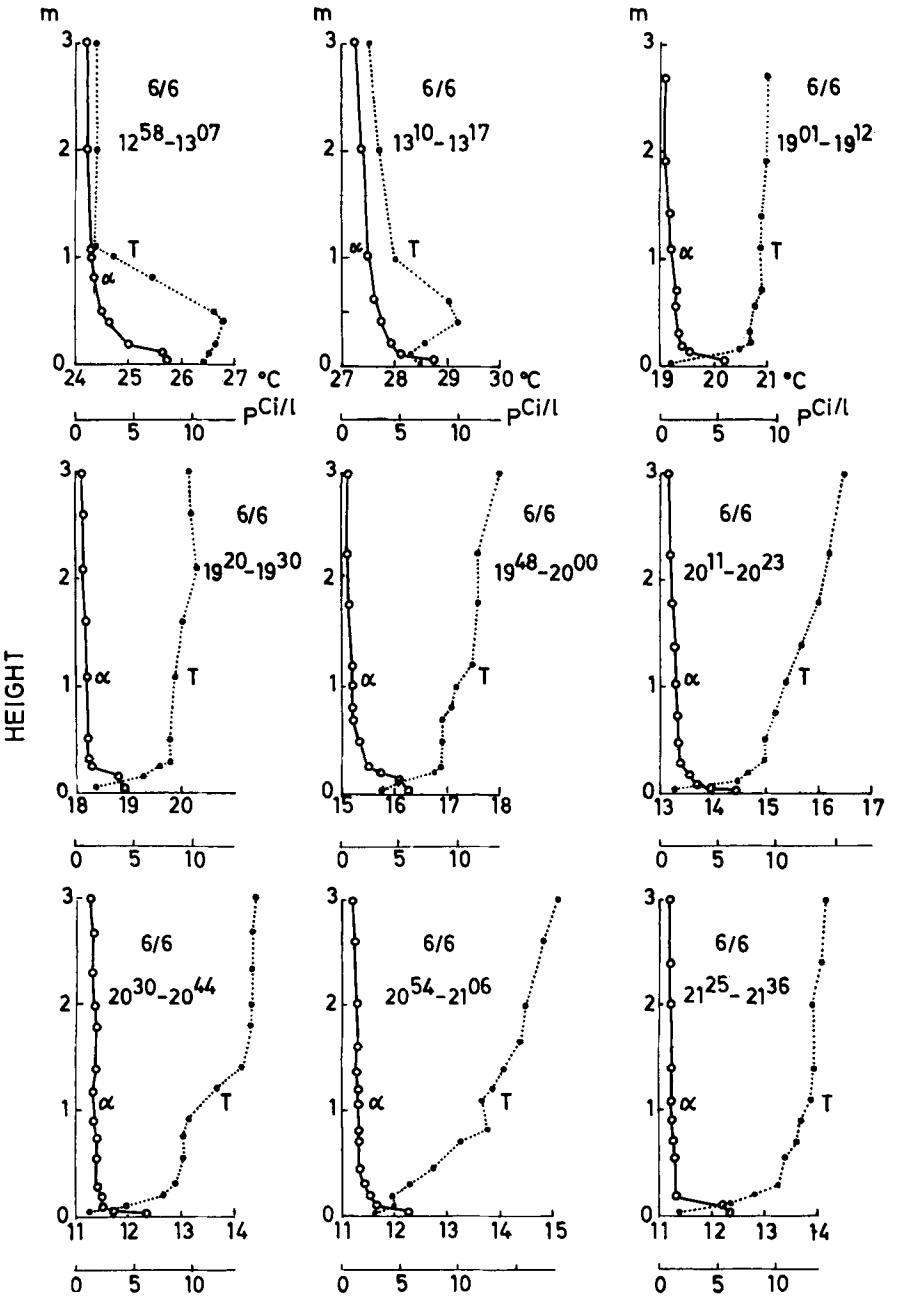

Fig. 9. Profiles of α-activity and temperature.

the Monin–Obukhov "similarity theory", there exist near the ground a velocity u^*, a length L, and a temperature T^* which are essentially invariant with height.

In the present investigation natural α-activity and temperature were recorded simultaneously in the 0–3 m layer. The copper vs constantan thermocouple was attached in the intake tube of the ionization chamber. Consequently the ventilation of the thermocouple was good.

The profiles of temperature and α-activity

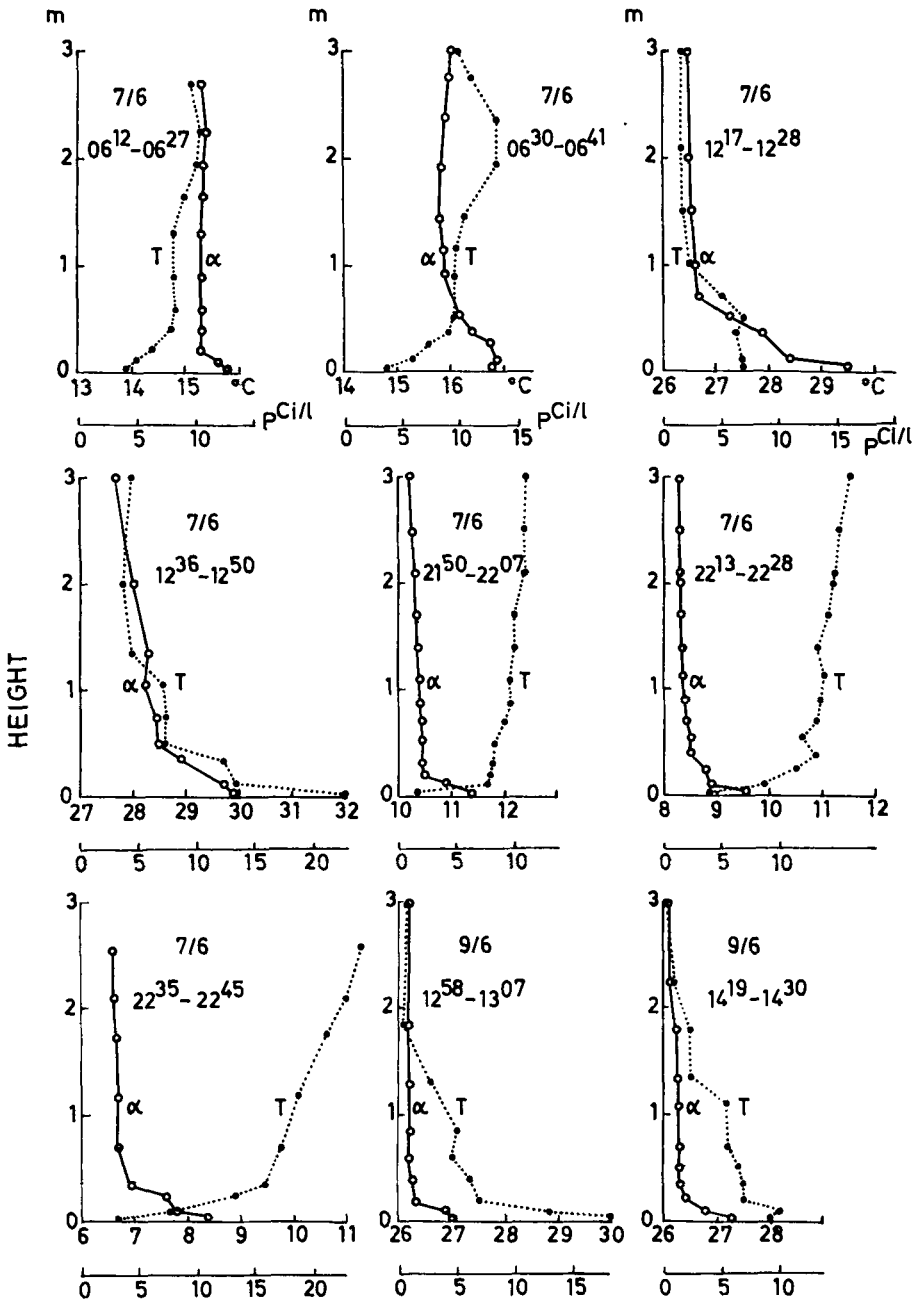

Fig. 10. Profiles of α-activity and temperature.

were obtained by gradually lifting the intake tube from the ground up to 3 m above the surface. The measurement period refers to clear days during summer time (June 6–7 and 9, 1970) with strong daytime insolation. Our preliminary data ought to give information on the structure of vertical profile of natural α-activity and on its variation with the stability of the lower layer of the atmosphere.

Figs. 9 and 10 show profiles of the natural

α-activity and temperature in the layer below 3 m. Rings mark α-activity; dots, temperature. The measurements nearest the ground refer to the height 0–5 cm due to the design of the intake tube. All the profiles of natural α-activity show a decrease with height with a maximum gradient close to the ground. The temperature profiles, however, show a less uniform variation due to the long-wave and short-wave radiation conditions. During daytime there is usually a decrease in temperature with height; at night, an increase with height.

The wind conditions were almost the same every day. The wind speed at 10 m above the ground varied between 3 and 4 m/s during daytime and between 2 and 4 m/s at night; the wind was northerly. This is especially important due to the fact that the exhalation rate of thoron may vary considerably with the area surrounding the observatory.

No closer investigations of such local variations have, however, been made as yet.

All profiles show that situations with a marked vertical variation of temperature are characterized by variation of natural α-activity. The profiles of natural α-activity also show that the gradients are not always greatest close to the ground. Thus in several cases the gradient was found to increase with height. Finally, the vertical gradient of α-activity does not seem to be very pronounced and the profiles of natural α-activity often show a smoothed structure. This is not the case with the temperature profiles.

The diagrams also show that the intensity of α-activity near the ground seems to be greater in daytime than during the night.

During daytime on June 6th, 1970, the profiles show a decrease in temperature with height above 40 cm, but near the ground there was an increase in temperature with height. On June 7th, between 12 and 1 o'clock, however, with a clear sky and a strong insolation, the temperature profiles show a more "normal" decrease with height from the ground surface and up to 3 m. All simultaneous α-activity profiles then show a decrease in activity with height.

The diagrams referring to the 9th of June show profiles which are similar to those mentioned above.

After sunset (7 to 11 o'clock) our measurements showed a pronounced temperature gradient near the ground, and the profiles were found to be almost linear in the lowest 30 cm. Above this level there was a less pronounced temperature rise with height. The α-activity profiles have maximum gradients at those heights where the greatest temperature gradients were observed. The profiles of 7th June. 6^h–7^h, show the vertical distribution of the natural α-activity and temperature after sunrise. In spite of a general temperature rise of one degree between 6 and 7 o'clock caused by the insolation we obtain the lowest values near the ground. The wind speed at the 10 m level was on this occasion about 3 m/s. The concentration of α-activity was fairly high, greater than 10 pCi/l. The high value is probably due to an accumulation of radon in the surface layer under inversion conditions during the clear night.

The irregularities which can be seen in some of our temperature profiles may be explained by the inhomogenity of the surface within the observatory area. Our observations were above a lawn with short grass and the adjacent areas were free from any vegetation thus creating different temperature conditions in the lower layers.

In the future mainly the radon and thoron profiles will be measured at Marsta Observatory.

Summary and conclusions

The aim of the present paper has been to present a new principle for measuring the variation with time of the natural radioactivity in the air near the ground. Especially the rapid fluctuations of the α-activity near the ground will be studied.

In order to achieve rapid, continuous measurements of the α-activity in air an aspiration method was developed similar to that given by H. Israël & G. H. Israël (1966). After passing through a filter that holds back all aerosol particles the air passed through a 24 litres ionization chamber. The ionization current was continuously registrated by a high-speed recorder. The time constant of the measuring equipment was approximately 6 sec.

A number of formulae previously given for the usual meteorological parameters have

been applied to the natural α-activity in the air near the ground. The preliminary results presented in this paper show an obvious connection between the fluctuations of natural α-activity and the temperature. The correlograms of α-activity and temperature seem to indicate some connecting link, apparently the vertical motion. The structure of the variation with time of the α-activity will be made more penetratingly in later papers.

In the present investigation we have also made the simultaneous measurements of the vertical structure of the natural α-activity concentration and the temperature in the layer up to 3 m above the ground. For all stability conditions we obtained a smoothed, decreasing concentration of the α-activity with height. The gradients were fairly great near the ground and decreased rapidly with the height. The temperature profiles, however, showed a less uniform structure, especially with stable stratification.

Acknowledgements

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О ЕСТЕСТВЕННОЙ α-АКТИВНОСТИ ВОЗДУХА ВБЛИЗИ ПОВЕРХНОСТИ ЗЕМЛИ. ЧАСТЬ А. МЕТОД ИЗМЕРЕНИЙ И НЕКОТОРЫЕ ПРЕДВАРИТЕЛЬНЫЕ РЕЗУЛЬТАТЫ

Разработан метод, с помощью которого концентрация естественной α-активности воздуха вблизи земли может быть изучена в широких пределах ее изменений. Метод основывается на процессе аспирации. Аппаратура используется в основном для измерений быстрых флуктуаций α-активности воздуха. Предварительные результаты, представленные в этой статье, указывают на связь между этими флуктуациями и температурой, существующей, возможно, благодаря связующему звену — вертикальным движениям. Были произведены дальнейшие одновременные измерения естественной α-активности и температуры в слое до 3 метров высотой. Градиенты были довольно большими вблизи поверхности земли и быстро уменьшались с высотой.