Measurement of the diffusion of radon, thoron and their radioactive daughter products in the lower layers of the Earth's atmosphere

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ABSTRACT

We undertook the study of the diffusion of radon, thoron and their radioactive daughter products in the lower strata of the atmosphere in relation to the variations of certain meteorological parameters, with the object of a possible future application to the measurement of the pollution and the diffusion of the air. We give the results of measurements which we have taken between 0 and 30 metres in altitude for the study of the vertical distribution of natural radioactivity, and in different regions in order to define the influence of certain local parameters.

The study of vertical distribution is carried out at the National Meteorological Observatory in Magny-les-Hameaux (Seine-et-Oise). A tower 30 metres high permitted us to take air samples continuously at several heights above the ground. We measured simultaneously the concentrations of radon, thoron and the daughter products of these gases. The variations in the concentrations and in the radioactive equilibrium between gas and daughter products are related to the variations of meteorological parameters and, in particular, to the diffusitivity of the air.

Introduction

The existence of radioactive elements, suspended in the air, certain of which can be easily identified, suggests their use as tracers for the study of certain problems of atmospheric physics. Natural radioactivity originating in the earth's crust, i.e., isotopes of element 86 and their radioactive daughter products, which are gaseous elements or very fine particles, are diffused in the atmosphere as are the other constituents of the air. They can consequently be useful for the measurement of stability and turbulent diffusion.

With this objective in mind, we undertook a study of the diffusion of radon, thoron and their daughter products in relation to the variations of certain meteorological parameters. These elements, besides having the above mentioned properties, also have the advantage of common origin, i.e., the ground, and very different radioactive periods. Radon has a half-life of 3.8 days: it gives birth to elements which can be grouped approximately into two groups: short-lived elements with apparent half-lives of 40 minutes. The latter in disintegrating give long-lived elements, RaD and RaF, the respective half-lives of which are 22 years and 140 days. Thoron, which is an isotope of radon, has a half-life of 54 seconds. Its daughter products have a half-life of 11 hours.

The concentrations of these gases or particles vary differently according to the time since their formation and it is interesting to study the evolution of their concentrations in relation to the distance from the source of radon or thoron, i.e., in relation to the height above the ground.

1. Theory of the diffusion of natural radioactivity

Radon and thoron are formed in the ground. They are set free in the atmosphere where they bring about a series of radioactive daughter products which are rapidly fixed on the dispersoids of the air. The action of gravity on these particles imparts to them a sedimentation speed v. Moreover, certain atmospheric phenomena (rain, snow etc.) bring about the precipitation to the ground of a part of the aerosols. One calls λ the wash-out constant which can be considered identical for the various natural

radioactive elements independent of the altitude, at least in the lower layers.

The diffusion of gas and different daughter products is generally represented by this equation for an element i with a concentration Q_i:

$$\frac{\partial Q_i}{\partial t} + U_x \frac{\partial Q_i}{\partial x} + U_y \frac{\partial Q_i}{\partial y} + U_z \frac{\partial Q_i}{\partial z}$$

$$= \frac{\partial}{\partial x}\left(k_x \frac{\partial Q_i}{\partial y}\right) + \frac{\partial}{\partial y}\left(k_y \frac{\partial Q_i}{\partial y}\right) + \frac{\partial}{\partial z}\left(k_z \frac{\partial Q_i}{\partial z}\right)$$

$$+ \lambda_{i-1} Q_{i-1} - (\lambda_i + \lambda) Q_i + v \frac{\partial Q_i}{\partial z}.$$

U_x, U_y, U_z, are the 3 components of the wind.
k_x, k_y, k_z, are the coefficients of diffusion according to the three components x, y and z.
λ_i is the radioactive constant of the itn daughter product.
z represents the altitude above the ground.

Some hypotheses make it possible to simplify this equation:

(1) One takes x parallel to the average wind direction.

(2) v being of the order of 1 m/h for particles of the order of a micron, the term $v(\partial Q_i/\partial z)$ can be neglected.

(3) One examines the phenomena in steady state, i.e., $\partial Q_i/\partial t = 0$.

(4) The emanation above the ground is taken as uniform in the x, y, plane, there is no horizontal gradient

$$\frac{\partial Q_i}{\partial y} = \frac{\partial Q_i}{\partial x} = 0;$$

we thus obtain the following system of differential equations.

$$\frac{d}{dz}\left(k_z \frac{dQ_1}{dz}\right) - \lambda_1 Q_1 = 0$$

for the radioactive gases; and for the daughter products:

$$\frac{d}{dz}\left(k_z \frac{dQ_i}{dz}\right) \lambda_{i-1} Q_{i-1} - (\lambda_i + \lambda) Q_i = 0, \quad i = 2, 3 \ldots$$

Two important points complicate the search for the solution: The definition of conditions at the limits and the form of variation of k_z with the altitude.

Conditions at the limits

For the gases, the conditions at the limits are easy to establish. The flux which crosses the surface of the ground is E_1.

$$\left(k_z \frac{dQ_1}{dz}\right)_{z=0} = -E_1$$

$$Q_1 \to 0 \quad \textit{for} \quad z \to \infty;$$

for the daughter products, one still has

$$Q_i \to 0 \quad \textit{for} \quad z \to \infty.$$

The second condition is more difficult to determine. In fact, the aerosols reaching the ground have a quite large probability of settling there. Let P be this probability ($0 < P < 1$) from which it results that the flux to the ground of daughter products is negative. The simplest hypothesis in expressing this flux is to say that it is proportional to the aerosol concentration at ground level. In this case we can write:

$$\left(k_z \frac{dQ_i}{dz}\right)_{z=0} = P \alpha Q_i,$$

where α is a scale factor whose dimension is $L^{-2}T^{-1}$ and which has to be examined experimentally. As for P it seems probable that it is equal to 1.

Form of variation of k_z

Numerous hypotheses have been made regarding the form of variation of the diffusitivity with altitude. A first approximation consists in considering k_z constant in relation to the altitude, cf. HESS (1918), FONTAN (1964). Others have adopted different laws of variation, cf. SCHMIDT (1923), PRIEBSCH (1931), MALAKHOV (1959), JACOBI (1963). In particular, Jacobi has calculated numerically, for gases and different daughter products, the variations of concentration with altitude for different profiles of k_z.

We shall not go in detail into the various hypotheses and their justifications. It can be said that outside of the friction layer where k_z is quite large, the variation profile of diffusitivity is connected approximately with the profile of the wind through correlations such as those given by SUTTON (1960).

In the boundary layer, the local meteorological parameters have a great influence on the

profile of k_z. In one of the first stages of our study of the diffusion of the natural radioactivity of the air, we studied the relation between the distribution with altitude of concentrations of different natural radioactive elements and the meteorological parameters, thermal gradients, speed and direction of the wind, temperature and humidity and precipitation.

2. Measurement of the vertical distribution of natural radioactivity in the lower layers

From the earlier work on this same problem, we shall mention that of SISIGINA in the USSR (1964) who measured the distribution of radon between 0 and 300 metres altitude. Sisigina adsorbes radon on active carbon and measures the concentration gradients which permit him to define a series of air layers in which he determines an average diffusion coefficient.

SERVANT (1964) and SERVANT & TANAEVSKY (1961) measured the concentrations in daughter products of radon at different altitudes of the meteorological tower of Saclay and of the Eiffel tower. MOSES & LUCAS (1960) at the Argonne National Laboratory in the U.S.A. measured radon between 0 and 40 metres by adsorbing it on silica gel.

Measurements of the concentration of radon were made at greater altitudes with the aid of airplanes by several investigators (KIRICHENKO, 1962, in the U.S.S.R., MACHTA, 1960–1962, and WILKENING, 1956, in the U.S.A.

We, for our part, are interested in the boundary layer between 0 and 30 metres and we have measured simultaneously in a continuous manner the concentrations of radon, thoron and short-life daughter products of these two gases, at two heights above the ground. These measurements make it possible to examine the concentration variations of different elements at the two levels.

3. Description of the experimental device

The experiments took place at the Magny-les-Hameaux National Meteorological Observatory (Seine-et-Oise). The land is relatively flat with low obstacles on the ground. A 30 metres high meteorological tower made it possible to take samples at 1.5 and 30 metres above the ground. The devices taking the samples at the two altitudes are identical. We shall briefly go over the principle and the characteristics (15, 16).

(a) Measurement of the concentration of gases

The air which is to be analysed for gas concentration is cleared of all its atmospheric aerosols by filtering. It then goes into a disintegration chamber where a part of the gas decays. The daughter products thus formed are collected and their α and β activities are measured with two scintillation detectors placed above and below the filter. The daughter products of thoron are separated from those of radon by an electronic coincidence. The α particle of the ThC′ is emitted with a period of 0.3 microseconds. A coincidence selector with a resolving time of a microsecond makes it possible to eliminate the α particles emitted more than one microsecond after a β particle. The RaC′ of which the period is $1.6 \cdot 10^{-4}$ second is thus eliminated. The number of coincidences gives the activity due to thoron. The α activity makes it possible to deduce the concentration of radon in the air. These devices make it possible to detect $1.4 \cdot 10^{-15}$ Ci/l of radon and $5.5 \cdot 10^{-15}$ Ci/l of thoron. The latter value corresponds to a sampling time of 12 hours. For an integration time of 2 hours, the sensitivity is $2.5 \cdot 10^{-14}$ Ci/l.

(b) Measurement of the activity of atmospheric aerosols

Atmospheric aerosols are collected directly on a fiber filter. The activity due to the daughter products of thoron is separated from that due to the daughter products of radon as in the preceding case. The sensitivities are 10^{-16} Ci/l for the daughter products of radon and $4 \cdot 10^{-17}$ Ci/l for the daughter products of thoron. This value is not more than $2 \cdot 10^{-16}$ Ci/l for a sampling time of two hours. Afterwards we shall design all the daughter products of Rn by RaB and RaC.

(c) Measurement of meteorological parameters

The temperature under shelter, the speed and direction of the wind, the ground evaporation, the relative humidity, and the thermal gradient between 20 and 30 metres are measured by National Meteorology Department.

4. Experimental results

The vertical distribution of elements constituting natural radioactivity is, as we have just

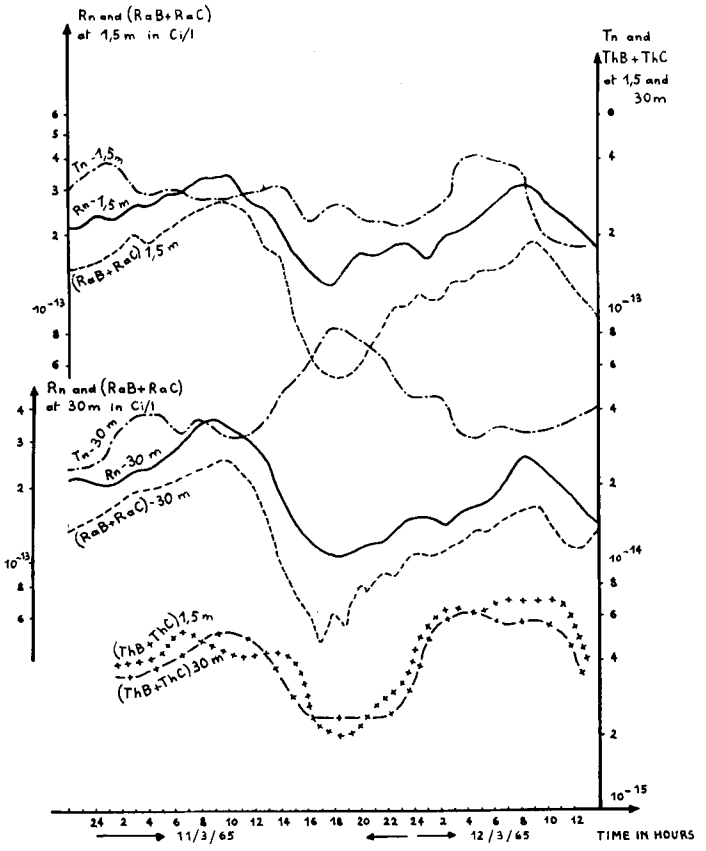

FIG. 1a

seen, a function of the diffusitivity of the air which is itself related to meteorological parameters. By means of examples we are going to show some relations between the local meteorological parameters characteristic of the stability of the atmosphere, and the concentrations which we have measured at different levels above the ground.

1. *Daily variations of the different elements*

In figure 1 (*a*, *b*) can be seen a frequent case of daily variations of the main elements and of their relations to the meteorological parameters. The thermal gradient is periodically positive, negative or zero, without ever reaching any large values. The similarity can be seen between the curves giving the concentration variations of radon and daughter products of radon and thoron, and those giving the meteorological parameters: relative humidity and thermal gradient. The temperature under shelter varies in the opposite way from the previous cases.

The concentrations of radon and of the daughter products of radon and of thoron are approximately the same at the two levels. The unbalance, greater at 1.5 metre than at 30 metres between radon and its daughter products, is low. Thoron having a shorter period shows different variations from those of the other radioactive elements. The concentrations which we are indicating are integrated over two or three hours. It is thus not possible for us to keep up with the rapid variations of the thoron concentrations. As shown by ISRAEL (1965) experimentally, and by JACOBI (1963) theoretically, the variations are different according to the altitude at which one makes the sampling. The ratio of the concentrations between 1.5 and 30 metres becomes lower as the turbulence becomes greater. Supposing a linear variation of k_z with the altitude between 1.5 and 30

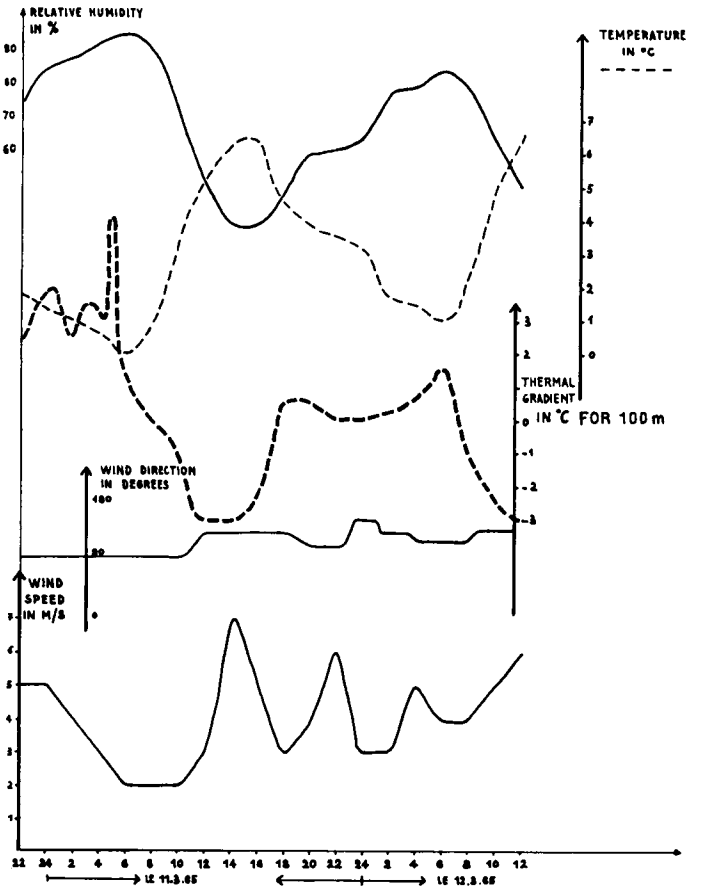
RELATIVE HUMIDITY IN %
TEMPERATURE IN °C
THERMAL GRADIENT IN °C FOR 100 m
WIND DIRECTION IN DEGREES
WIND SPEED IN M/S
LE 11.3.65
LE 12.3.65

FIG. 1b

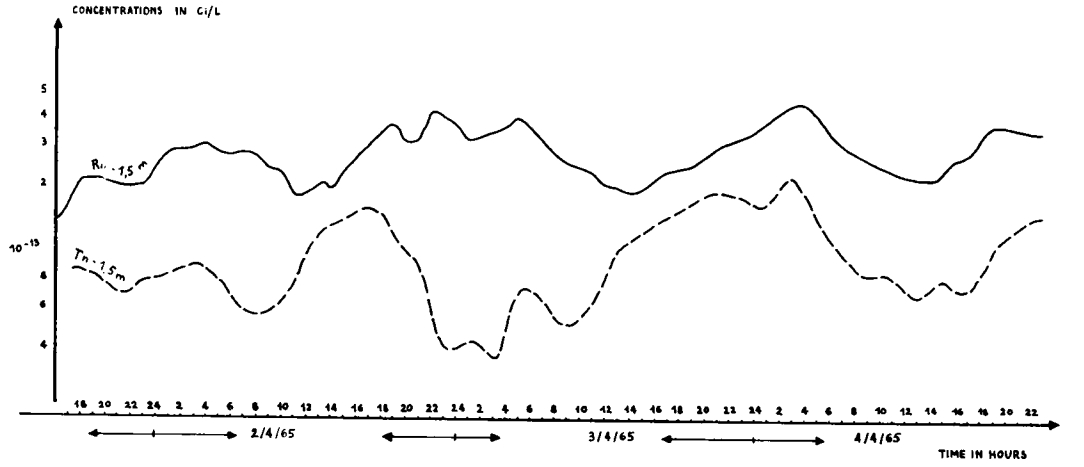
CONCENTRATIONS IN Ci/L
10⁻¹³
Rn - 1,5 m
Tn - 1,5 m
2/4/65
3/4/65
4/4/65
TIME IN HOURS

FIG. 2

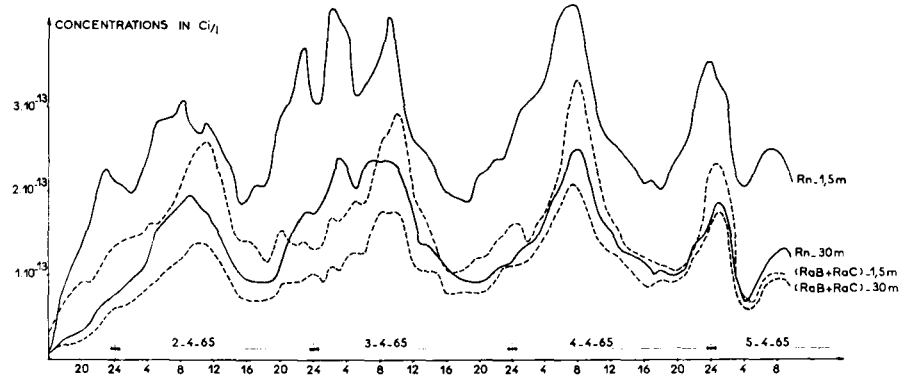

FIG. 3a

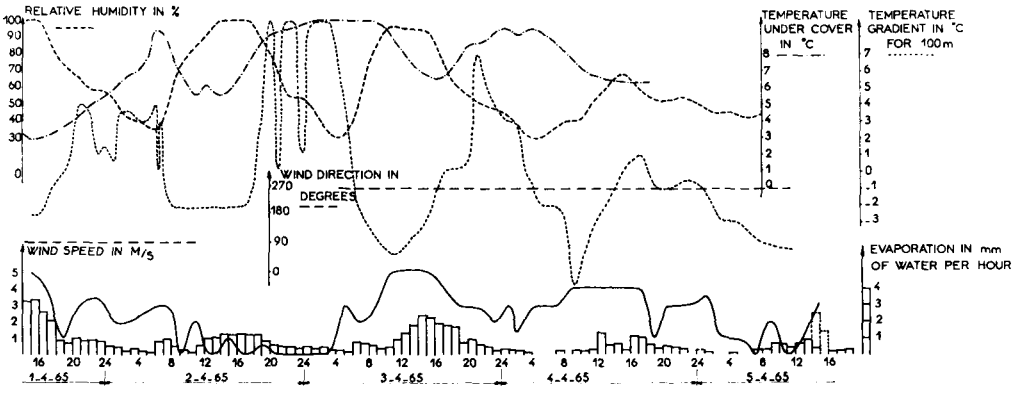

FIG. 3b

metres, the concentration ratio at these two heights above the ground indicates that at $z = 1.5$ metre, k_z varies between 1 and 10^{-1} m²/s throughout the day.

At 1.5 metre above the ground one observes a maximum followed by a decrease in the concentration of thoron while the stability of the atmosphere between 0 and 1.5 metre increases. In fact, when the atmosphere becomes very stable, thoron disintegrates before having diffused up to 1.5 metre. By assuming a linear variation of k_z between 1 cm and 1.5 metre, this maximum corresponds to k_z 10^{-2} m²/s at 1.5 metre.

Figure 2 shows the variations of radon and thoron compared at 1.5 metre. The concentration of thoron reaches a maximum and then diminishes, whereas the concentration of radon continues to increase, which shows clearly an increase in the stability of the air. Figure 3 (*a*, *b*) gives the corresponding recordings of radon and its daughter products at 30 metres and the meteorological parameters.

2. *Temperature inversions located at ground level*

We have just seen in the preceding example the thermal gradient inversion shows itself as an increase in the concentration of radon and of its daughter products at 1.5 metre and 30 metres.

Temperature inversions are sometimes located at the level of the ground and the variations at 1.5 and 30 metres are then very different. Figure 4 shows such a phenomenon. The concentration variations of radon at 1.5 metre is accompanied by a decrease at 30 metres which clearly shows the absence of exchange between

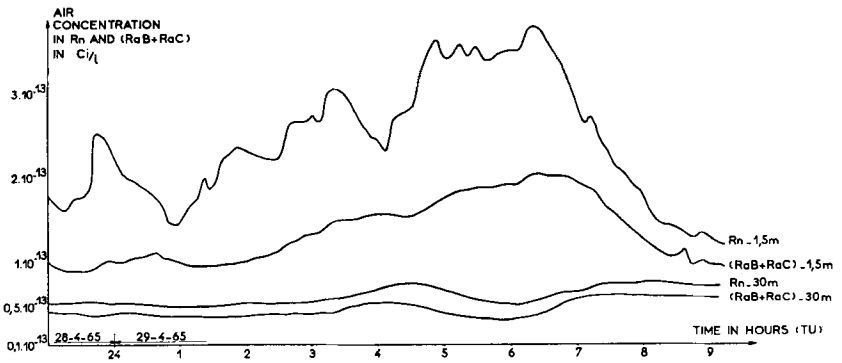

FIG. 4

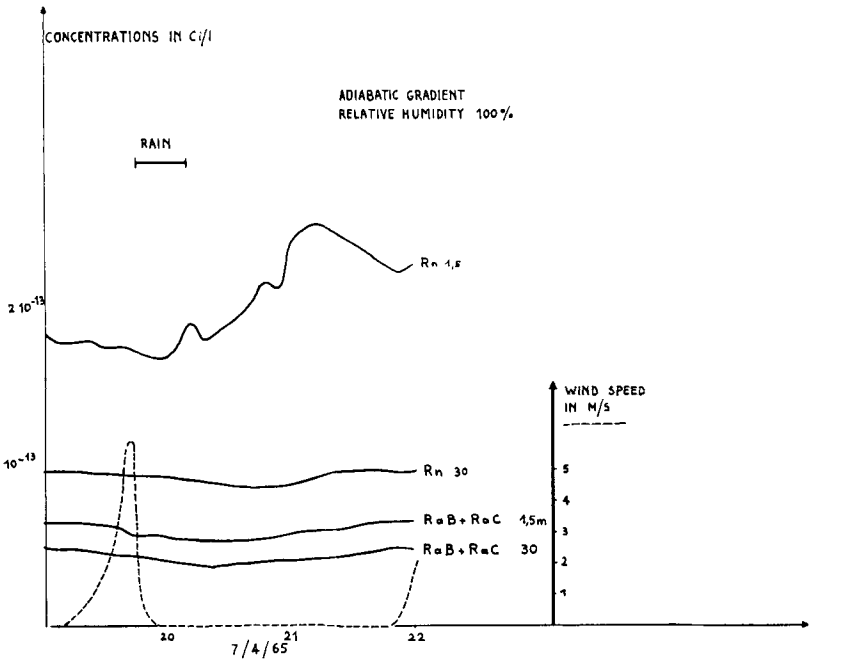

FIG. 5

the 1.5 and 30 levels. The decrease in the concentration at 1.5 metre is followed by an increase at 30 metres, which corresponds to the movement of the inversion layer upwards. The ratio between the concentrations at 1.5 and 30 metres can reach a value of 10. The inversion phenomenon is expressed at ground level by an unbalance between radon and its daughter products. It is due to the fact that the daughter products lack the time to attain equilibrium with radon during rapid variations, and also to the proportion of daughter products which are deposited on the ground. The influence of the latter is very great, the layer where the accumulation takes place not being very thick.

These inversions are frequently produced after rain or snow which brings about a cooling of the ground. In figure 5, we can see the increase of the radon concentration at 1.5 metre level after a rain. In figure 6 the same phenomenon is shown after a snow-fall. There is an increase at ground level of the radon concentration and a decrease in the concentration of daughter products. We attribute these varia-

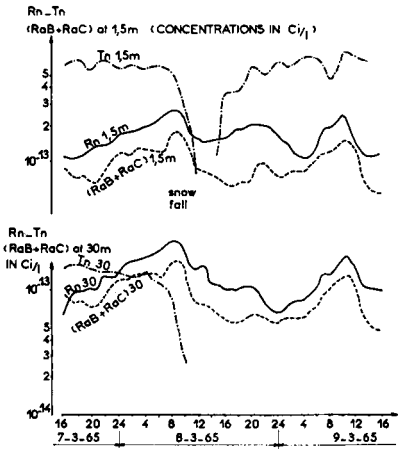

FIG. 6

tions to the influence of the ground which captures the daughter products, the inversion layer being very thin.

3. *Influence of precipitation*

Precipitation in general does not bring about a variation in the ratio between gases and daughter products. We have observed such a variation only in the case of a fine rain or drizzle (3). This means that the rain has only a slight filtrating power for particles with dimensions of the order of 0.1 micron. This finding is moreover in agreement with those of other investigators who have shown that the radioactivity of the rain was fixed on the drops mainly at the cloud level and at the moment of formation.

Precipitations brings about a decrease in the emanation of thoron from the ground. After a long period of rain, followed by periods of sunshine, one observes a slow increase in the concentration average of thoron at the two levels.

Figure 6 shows the influence of snow-fall on the release of thoron. The ground being covered with snow, a rapid decrease takes place in the concentration of thoron at the two altitudes 1.5 and 30 metres.

This phenomenon is more difficult to observe in the case of radon due to its longer half-life. Even in the absence of release, its concentration in the air decreases only slowly.

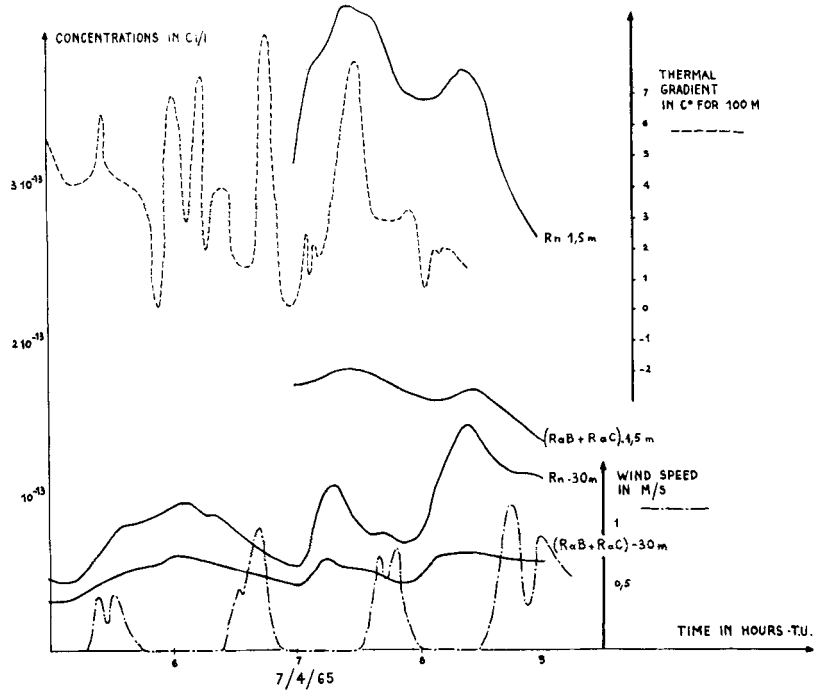

FIG. 7

4. *Influence of the winds*

The number of measurements which we have made at Magny-les-Hameaux is still not sufficient to permit us to set up a local relationship between natural radioactivity and the direction of the wind. Magny-les-Hameaux is located in the Paris region, quite a way in the interior, and far from the Atlantic Ocean. The sea air has thus had the time to become charged with radon. The wind comes in as a generator of turbulence. There is evidently a relationship between the direction and the speed of the wind, but we think that it would first of all be interesting to establish the correlation between the thermal gradient on the one hand, and the speed and direction of the wind on the other. The temperature inversions take place mainly in continental air masses. One observes in this case a strong increase in natural radioactivity.

Figure 7 shows the influence of local winds. They are accompanied by rapid variations in the concentrations of radon, but also in the stability of the air. Each gust of wind corresponds to a concentration decrease of radon, mainly at 1.5 metres, and of the thermal gradient (which is positive).

Conclusion

The first results obtained by measuring the vertical distribution of natural radioactivity in the lower layers of the atmosphere, show that the concentration of the different elements is strictly related to the stability of the air.

Natural radioactivity should permit the measurement of atmospheric diffusion. Thoron, because of its very short period, is very interesting for the study of the first few metres above the ground. Radon and the daughter products of radon and thoron can make possible the study of exchanges up to altitudes higher than the tropopause.

Certain points still have to be determined, such as the rapid measurement of radon and thoron flux from the ground without any modification of the release, the number of particles which are caught by the ground, the irregularity of ground release and the theoretical study in the transient state.

The existing installation will be soon complemented by the addition of an ionization chamber for the instantaneous measurement of thoron (ISRAEL, 1965), and from the meteorological point of view, by measurements of the profile of the wind and the temperature gradient between 1.5 and 30 metres.

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ИЗМЕРЕНИЕ ДИФФУЗИИ РАДОНА (Rn^{220} И Rn^{222}) И ПРОДУКТОВ ЕГО РАСПАДА В НИЖНИХ СЛОЯХ АТМОСФЕРЫ ЗЕМЛИ

Изучается диффузия радона (Rn^{220} и Rn^{222}) и продуктов его радиоактивного распада в нижних слоях атмосферы и связь ее с изменением метеорологических параметров с целью применения в будущем для измерения загрязнения и диффузии воздуха. Приводятся результаты измерений радиоактивности выполненных в различных местах для определения влияния местных метеорологических параметров, и распределение естественной радиоактивности по высоте в слое 0 до 30 метров выполнены в национальной метеорологической обсерватории в Magny-les-Hameaux (Seine-et-Oise). Наличие здесь 30-метровой башни позволяло брать пробы воздуха непосредственно на различной высоте над поверхностью земли. Во взятых пробах, немедленно, измерялась концентрация радона и продуктов его распада. Изменения концентрации и радиоактивного равновесия между газом и продуктами распада связывалось с изменениями метеорологических параметров, в частности, с диффузией воздуха.