Temporal and spatial variations of the concentration of the short-lived decay products of radon in the lower atmosphere

By J. SERVANT, *Centre National de la Recherche Scientifique Gif-sur-Yvette (S.- & -O.), France*

(Manuscript received October 25, 1965)

ABSTRACT

The study of natural radioactivity in the lower atmosphere will provide information on turbulent diffusion and transport of air masses in the lower atmosphere. We have, therefore, undertaken a two-part research program.

Firstly, we have made a study of natural radioactivity of the atmosphere above a plateau and, secondly, we have taken measurements at different latitudes and longitudes over the continents and the oceans, to obtain the general distribution of the radioactivity in the lower atmosphere.

We have found that the radioactivity of the air is much more important over the plateau than over the oceans (100 and 6×10^{-12} Ci m^{-3}, respectively) and more important over the oceans in the Northern Hemisphere than in the Southern Hemisphere (6 and 1.5×10^{-12} Ci m^{-3}, respectively). From these results, we can suppose that it will be possible, with the aid of natural radioactivity, to identify air masses and to follow the vertical and horizontal diffusions in a given mass of air.

Some decades ago, the variations in the composition of the atmosphere were studied only by meteorologists and physicists. Today, this problem has become popular. Everyone asks about the substances that he breathes and their concentrations. To give an answer to this last point, we must study in particular the air movements in the lower atmosphere. Many workers have shown that radon is a convenient tracer for the study of movements of air masses [B1, F1, S2] and of the atmospheric stability [A1, F1, I1, J1, M1, S1, S2, T1, T2, W2]. Some of them have investigated the radioactive source—i.e. the flux of radon and its variations with the geological characteristics of the soil [P1, S2, S3, W1]; others, the radioactivity of the air with regard to the stability of the atmosphere; but few have shown the influence of different geographical conditions, and hence we have not a precise idea of the importance of these factors in determining the radioactivity of the air at a given site.

In this paper, we shall present the information that we have obtained with radon, continuously given off by the soil, (I) above a plateau, (II) above a hilly site [S1], and (III) above the oceans. The geographical, geological and dynamical conditions are very different in these three sites and allow us to appreciate how these factors influence the variations of the radioactivity of the air.

I. Study of the radioactivity of the atmosphere above a plateau

The site chosen is the plateau of Saclay. It is located in the boundary of the Parisian region and it spreads over several dozens of kilometers of a sedimentary soil. This site is particularly favourable for the study of the influence of different dynamical conditions on the radioactivity of the air as it represents a flat horizontal area with a uniform flux of radon throughout the year. The air analysis was made between 0 and 100 meters from a meteorological tower and radioactivity measured was that due to short lived daughter products of radon. The radioactive aerosols of the air were retained on a filter paper and the α activity measured instantaneously with a scintillation counter [S2].

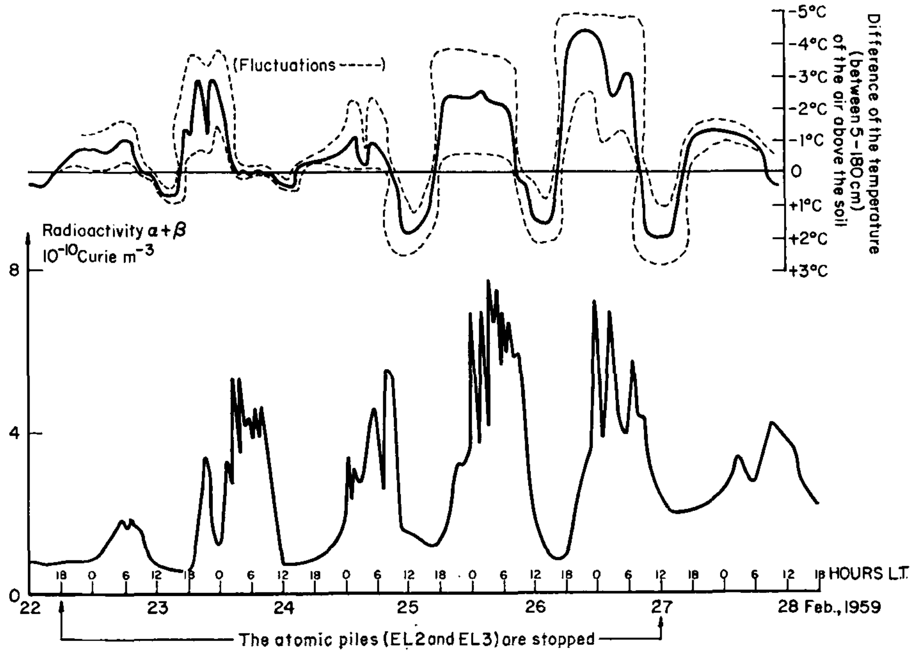

FIG. 1. Variations of the natural radioactivity, and of the difference of temperature of the air above a plateau.

General observations

In general, we have observed a succession of weak and strong concentrations of the active deposit over 24 hour periods, which is in connection with stability variations of the atmosphere due to the solar radiation, and the nocturnal radiation of the earth surface. The very high radioactivity was observed during the night when an inversion of the air temperature was detectable near the soil.

Near the soil, a typical example is given in the figure 1. The inversion of the air temperature generally occurred only during the night. If this inversion persisted during the day, the radioactivity of the air remained high, as illustrated in figure 2 (7th, 8th and 9th February, 1960). A condition which must also hold is a wind of moderate velocity near the soil (horizontal speed less than 0.5 ms^{-1}); in the region considered, the wind was blowing from N.-NE.

From 0 to 100 meters, the following recordings have been obtained (figure 3): during nights with a greater atmospheric stability, the radon emitted from the soil accumulates in a layer to the height of 30 meters (9th, 10th, 11th September, 1961). After sunrise at 06.00 hr, the vertical transport of radon increases, whereas near the soil the atmospheric radioactivity begins to decrease and this decrease continues throughout the day. At 09.00 hr, an increase of the air radioactivity can be observed at 100 meters; this is due to the passage of the air which had stagnated near the soil during the night (the ascending speed is equal to 1 cm s^{-1}). It has to be noticed that the tendency to atmospheric stability is obtained by Richardson's number as well as by the quantity of radon which accumulates in the lower atmosphere.

Particular observations

Usually, changes in natural radioactivity were slow and continual, but, sometimes during the night, rapid variations were observed (figure 4). For example, during the night of the 2nd to the 3rd October, 1961, considerable variations were recorded in radioactivity near the soil at 23.00 hr, 03.00 hr and 05.00 hr. The observation of the quantity of the active deposit in the different columns of air allows us to affirm that the column which passes between 06.00 hr and 08.00 hr has been submitted to certain

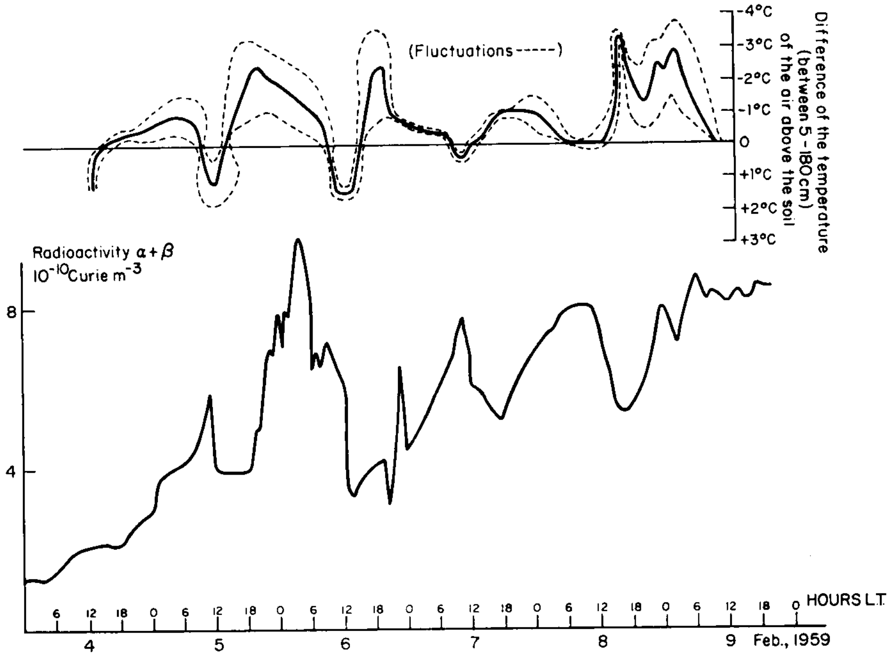

FIG. 2. Increase of the natural radioactivity of the air with an inversion of the air temperature above a plateau.

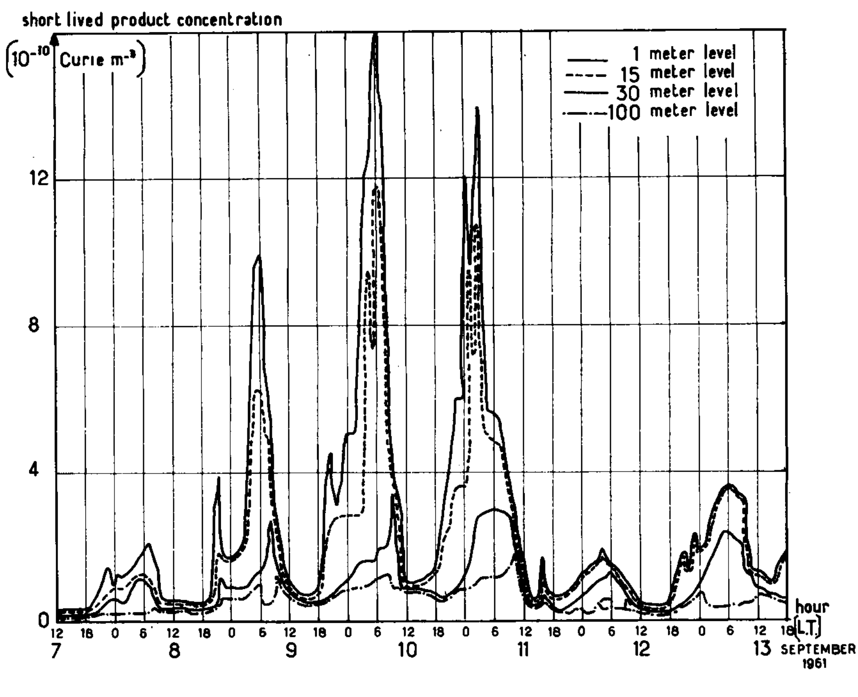

FIG. 3. Natural radioactivity of the air above a plateau (0–100 meters).

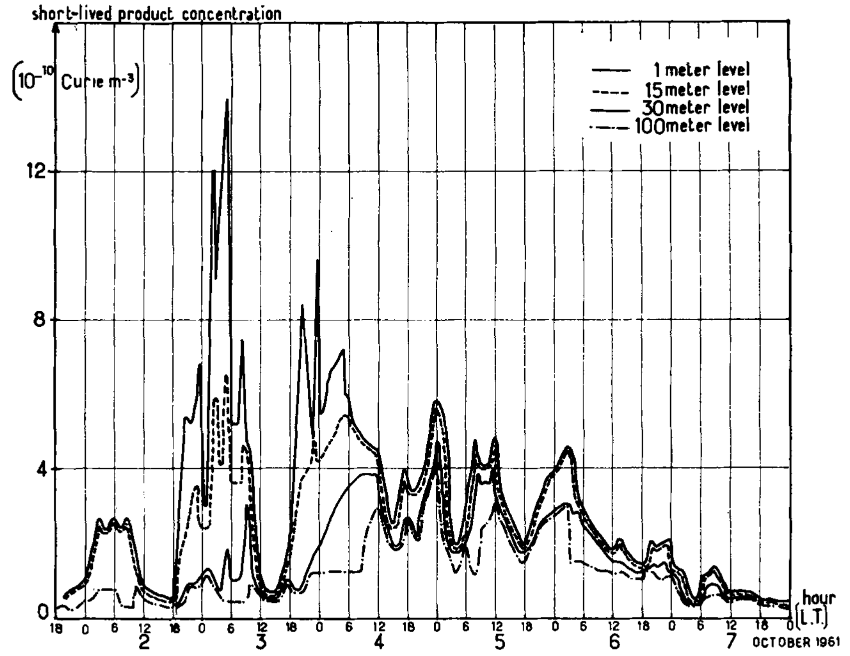

FIG. 4. Natural radioactivity of the air above a plateau (great variations during the nights).

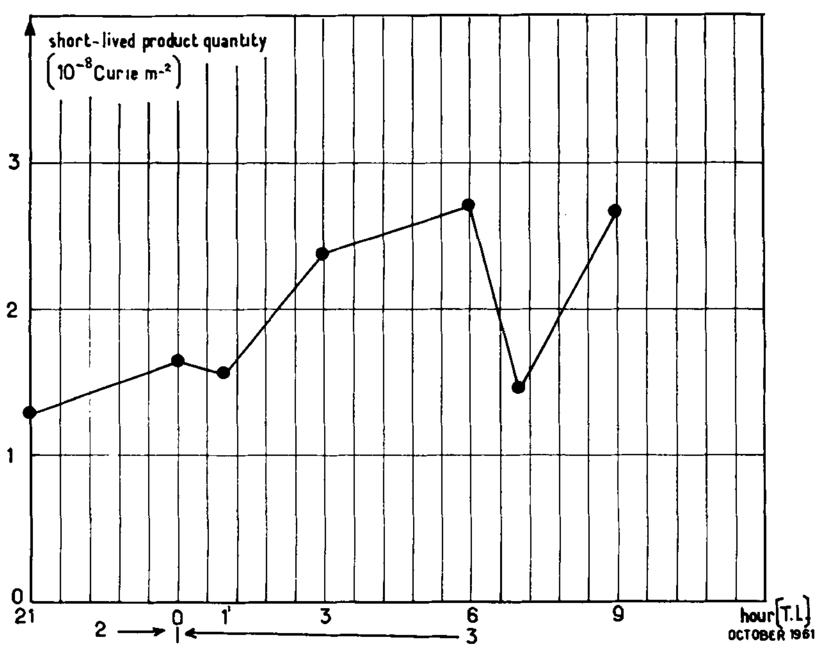

FIG. 5. Short-lived product quantity in the air columns.

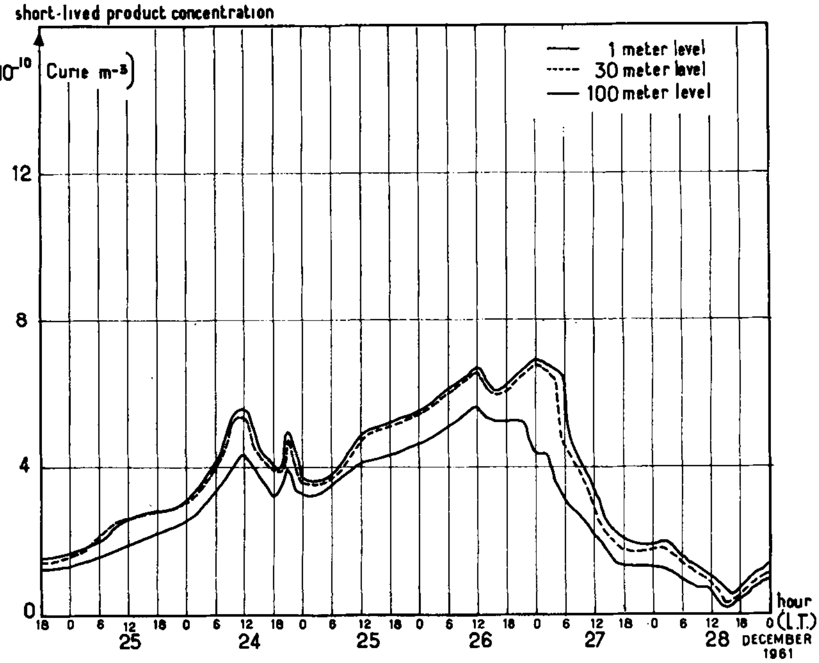

FIG. 6. Accumulation of the short-lived products of radon in the lower atmosphere.

conditions of stability different from the other columns (it has the lowest radioactivity (figure 5) and exhibits no increase in its radioactivity at 100 meters of altitude, moreover the sharp diminution of the activity at 30 meters is not usual at sunrise).

Another example of particular conditions is given in figure 6. During certain other days, the radioactivity continually increased near the soil and it was nearly constant throughout the column (0 at 100 meters).

CONCLUSIONS

1. The active deposit and radon can be used to study the stability of the atmosphere.
2. An air mass characterized by a particular temperature and relative humidity, cannot be recognized by its radioactivity when it passes on at air station, the night being sufficient for the loss of its identity.

3. *Flux of radon at a given site*

From the observed quantity of active deposit accumulated in the lower atmosphere during a clear sky night we can compute this flux; for example, at Saclay the following results have been obtained (number of observations in brackets):

1.55, 10^4 at. m^{-2} s^{-1}	July 1961 (9)
1.31, 10^4 at. m^{-2} s^{-1}	Sept. 1961 (5)
1.00, 10^4 at. m^{-2} s^{-1}	Oct. 1961 (3)

In 1958, the values obtained with iron-bells (area = 1 m^2, capacity = 100 liters) [S2] were 0.6 to 0.7 time lower than those given by the analysis of the atmosphere. Our result is comparable with the universal mean computed by H. Israel which is equal to 7.4 10^3 at. m^{-2} s^{-1}. This method has the advantage of the possibility of analysis of the degagement by a large surface (some hundred square kilometers) without any modification.

4. *Rate of renewal of air in the lower atmosphere*

The rate of air renewal has been easily determined in the lower atmosphere by measurements during a day following a night of high air radioactivity. For this purpose, the quantity of radon has been measured at the beginning and at the end of this period and the rate of

TABLE 1. *Rate of air renewal in the lower atmosphere.*

Interval of time (hr)...	6–9	9–12	12–15	15–18
Mean	0.42	0.60	0.59	0.61
Range (min.–max.)	0.18–0.71	0.38–0.74	0.29–0.87	0.50–0.85

renewal has been computed from the following expression:

$$\frac{Q \text{ initial} + Q \text{ escaped} - Q \text{ final}}{Q \text{ initial} + Q \text{ escaped}}$$

The air above 100 meters is assumed to be radon free and the radon flux is taken as equal to that of the previous night.

The calculations for the sunny days of July 1961 are given in Table 1. On an average, the rate of renewal is low from 06.00 hr to 09.00 hr, then, it becomes rapid and nearly constant. However, considerable variations appear within a given period, and this fact is evident from the maximum and minimum values. From 06.00 hr to 18.00 hr, 96 per cent of the air of the lower atmosphere is renewed by the air coming from layers higher than 100 meters.

5. Stability of various layers of the lower atmosphere

The quantity of radon which stagnates in the lower atmosphere can be used to determine the stability of the air in this layer. During clear sky nights, 9th, 10th, 11th September, 1961 when the stability was high, the quantity Ra A + Ra B + Ra C + Ra C′ + Ra C″ was five times that of cloudy nights (e.g. September 8th, 1961) and 18 times that of days with high turbulence (e.g. September 7th, 1961).

In the same manner, it is possible to give more details concerning the stability of various layers: during a period of stability, 75 per cent of the radon content of the lower atmosphere is accumulated in a layer at a height lower than 30 meters, whereas during a period of strong turbulence this ratio is only 30 per cent.

6. Measurement of the coefficient of vertical turbulent diffusion

Supposing that at a given point of the plateau, the degagement of radon of the soil occurs at a constant rate and that the turbulent diffusion coefficient does not change with the altitude, we can calculate a mean coefficient. For the period of September, from the 7th to the 13th, 1961, the following results were obtained (see Table 2):

The coefficient varies from 10^2 to 10^6 cm^2 s^{-1}. Lower values are observed between 21.00 hr and 09.00 hr (between 10^2 cm^2 s^{-1} and 2.10^4 cm^2 s^{-1}), and higher values between 09.00 hr and 21.00 hr (between 7.10^2 cm^2 s^{-1} and 10^6 cm^2 s^{-1}). The values lower than 10^3 cm^2 s^{-1} are attained only during the nights when the stability of the atmosphere is remarkable (9th, 10th, 11th September, 1961).

II. Study of the radioactivity of the atmosphere at a hilly site

This site is located in the Morvan region. The observations have been made near the soil

TABLE 2. *Mean coefficient of vertical turbulent diffusion in the lower atmosphere.*

Time (L.T.), hr...	18–21	21–0	0–3	3–6	6–9	9–12	12–15	15–18
Date (Sept. 1961)								
7							$4\cdot10^5$	$4\cdot10^5$
8	$4\cdot10^4$	10^4	10^4	$4\cdot10^3$	$6\cdot10^3$	10^5	10^5	10^5
9	$8\cdot10^2$	$2\cdot10^4$	$2\cdot10^3$	$2\cdot10^2$	10^4	$6\cdot10^4$	$2\cdot10^5$	$9\cdot10^4$
10	$7\cdot10^2$	10^3	10^3	10^2	$7\cdot10^3$	10^5	10^5	$2\cdot10^4$
11	$2\cdot10^3$	10^3	$3\cdot10^2$	$3\cdot10^3$	$5\cdot10^3$	$3\cdot10^4$	10^6	10^5
12	$9\cdot10^4$	10^4	$8\cdot10^3$	$2\cdot10^4$	$2\cdot10^4$	10^5	$9\cdot10^4$	$9\cdot10^4$
13	$8\cdot10^3$	$4\cdot10^3$	$4\cdot10^3$	$3\cdot10^3$	$3\cdot10^3$	$4\cdot10^4$	$5\cdot10^4$	$7\cdot10^3$

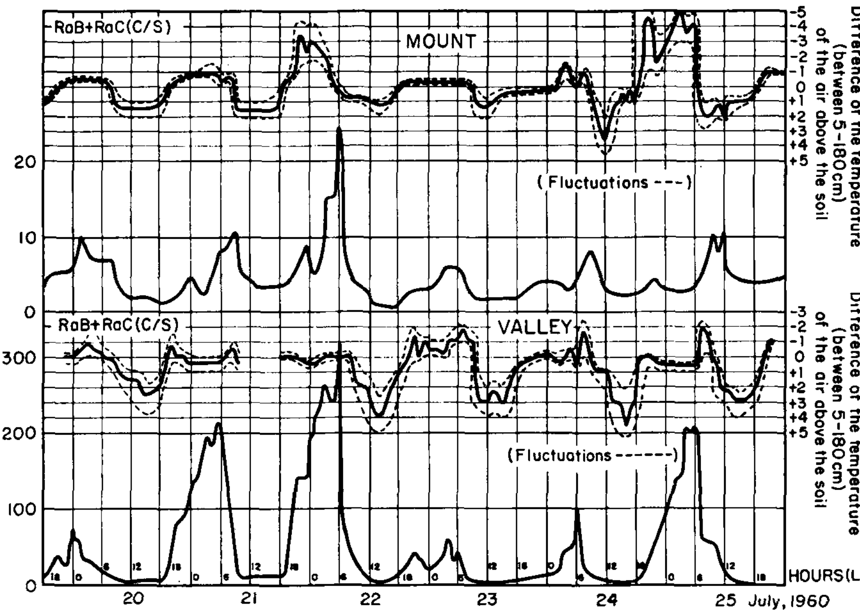

FIG. 7. Natural radioactivity and difference of the air temperature in a valley and on a mount.

in a valley and on an isolated mount. These two stations of measurements are separated by a distance of four kilometers and the difference of altitude is 320 meters. This site differs from the plateau of Saclay in its geography and its granitic soil. We have studied the influence of these variations in geological and geographical conditions and an example of the observations recorded is given in figure 7.

We have observed that the radioactivity of the air in the valley is proportional to the radioactivity of the granitic rocks only at night, and then is ten or twenty times higher than over the mount. Hence, near the soil of the valley, the radioactivity is fifteen times higher than at Saclay, i.e. 7.5 10^{-8} Ci m^{-3} or five times the permissible dose for the population (1.5×10^{-8} Ci m^{-3}). During the day, the radioactivity has usually the same value in the valley, on the mount and at Saclay. We have also observed downslope breezes during the nights (21st, 22nd, 24th, 25th July, 1960) with a speed of 0.2 ms^{-1}. After the sunrise, the upslope breeze is marked by the increase of the radioactivity on the mount at 09.00 hr (21st, 26th, 25th July, 1960).

III. Study of the radioactivity of the air above the oceans (at sea level)

The results obtained over several years are given in the Table 3. The radioactivity of the

TABLE 3. *The activity of the air due to the short-lived daughter products of radon collected on filter papers.*

Place	Mean annual (10^{-12} Ci m^{-3})	Min.–Max. (10^{-12} Ci m^{-3})
Mediterranean Sea[a]	100	30–200
Red Sea + Aden gulf[b]	50	13–90
Indian Ocean[c]	2	0–9
N. Atlantic Ocean[d]	6	0–280
Caribbean Sea[e]	7	0.7–26
S. Pacific Ocean[f]	1.5	0.2–9.2
Tasmanian Sea[g]	3.7	0.5–15.8
Antarctic Ocean Terre Adélie[h]	0.8	0–4

[a] 4 measures in November 1964.
[b] 8 measures in November 1964.
[c] 2 years of measurement at the Kerguelen islands + 22 measures between 20° N and 50° S in December 1964.
[d] 3 years of measurement at point A, J, K (1959–1962) + 25 measures between 20° N and 45° S in December 1963–1964. (Measures made by G. Lambert.)
[e] 8 measures in December 1963 and December 1964.
[f] 42 measures in December 1963 and December 1964.
[g] 15 measures in December 1963 and December 1964.
[h] 3 years of measurement in Terre Adélie (1959–1962) and on the Ocean (1963–1964). (Measures made by G. Lambert and G. Polian.)

air at the surface of the oceans is 10 to 100 times lower than that above the continent (100×10^{-12} Ci m^{-3}) and about 2 to 6 times lower in the southern hemisphere than in the northern hemisphere. This activity is due partly to the diffusion of radon in the upper layers of the low air of the atmosphere (near the shores) and to the radioactive decrease of the radon during its travel above the oceans which have a flux of nearly zero. For example, we have noted that an air mass which started from the Kerguelen Islands with an activity of 1×10^{-12} Ci m^{-3} passed over Australia with an activity of 0.5×10^{-12} Ci m^{-3}; this decrease agrees with the time taken by the air mass to go from the first station of measurement to the second station. The fluctuations near the mean value are due to the passage of the air masses of different origins [F1] and of different initial radioactivity.

Conclusion

The concentration of radon in the air of the lower atmosphere around the earth shows very important variations. It attains its maximum, $75{,}000 \times 10^{-12}$ Ci m^{-3}, above the continents and its minimum, 10^{-12} Ci m^{-3}, above the oceans, differing by a factor of 7.5×10^4.

Above the continents, the radioactivity of the air is controlled by the local meteorological, geological and dynamical conditions and gives a picture of the rate of the accumulation or of the escape from the lower atmosphere of a substance released by the soil.

For example, above the plateau, the radioactivity of the air is in direct relation with the stability of the atmosphere; in a hilly site, placed on a geoanomalous area, the radioactivity is governed by the stability of the atmosphere by the duration of the passage of air above the anomalous region and by the geographical position (e.g. during the night, the increase of the radioactivity is high in the valley but low on the mount). In general, these results confirm the previous meteorological observations. However, they give new precisions, above all in determining the stability of the atmosphere.

Above the oceans, there are the general meteorological conditions which determine the level of this radioactivity which becomes a witness of the history of the turbulent diffusion at a mean and a large scale.

REFERENCES

A1. Ardouin, B., Jehanno, C., Labeyrie, J., Lambert, G., Tanaevsky, O., and Vassy, E., 1963, Résultats des mesures de concentration dans la basse atmosphère des aérosols radioactifs artificiels. Rapport C.E.A., n° 2261.

B1. Becker, F., 1934, Messungen des Emanationsgehaltes der Luft in Frankfurt a.M. und am Taunus-Observatorium. *Beitr. Geophys.*, **42**, pp. 365–384.

F1. Fontan, J., 1964, Le dosage des radioéléments gazeux donnant des produits radioactifs de filiation. Son application à la mesure de la radioactivité naturelle de l'atmosphère. Thèse n° 218 de la Faculté des Sciences de l'Université de Toulouse.

I1. Israël, G. W., 1965, Der Thorongehalt in der bodennahen Atmosphäre. Dissertation, Technischen Hochschule Aachen.

J1. Jacobi, W., 1963, The vertical distribution of Rn-222, Rn-220 and their decay products in the atmosphere. *J.G.R.*, **68**, pp. 3799–3814.

M1. Les isotopes radioactifs dans l'atmosphère et leur utilisation en météorologie. Moscou 1965.

M2. Moses, H., Stehney, A. F., and Lucas, H. F., Jr., 1960, The effect of meteorological variables upon the vertical and temporal distributions of atmospheric radon. *Journal of Geoph. Res.*, **65**, Nb 4, pp. 1223–1238.

P1. Pearson, J. E., 1965, Radon-222. A study of its emanation from soil, source strength, and use as a tracer. Research reports, Dept. of General Eng., Univ. of Illinois. WRBANA.

S1. Schmidt, W., 1926, Zur Verteilung radioaktiver Stoffe in der freien Luft. *Phys. Zeitschr.*, **27**, pp. 371–377.

S2. Servant, J., 1964, Le radon et ses dérivés à vie courte dans la basse atmosphère. Thèse de doctorat ès Sciences, Paris. Rapport C.E.A., R.2434.

S3. Sissigina, T. T., 1962, The measurement of the exhalation of radon from the surface of certain types of rocks. *Problems of Nuclear Meteorology*. Ed. by I. L. Karol and S. G. Malakhov, Moscow. AEC — tr. 6128 — TID 4500, 25th Ed., pp. 125–138.

T1. Tanaevsky, O., and Vassy, E., 1955, Variations de la radioactivité naturelle et artificielle de l'atmosphère. *C.R.*, **241**, pp. 38–40.

T2. Tanaevsky, O., and Vassy, E., Radioactivité naturelle et artificielle de l'atmosphère. *Annales de Géophysique*, **11**, Fs. 4, pp. 486–490.

W1. Wilkening, M. H., and Hand, J. E., 1960, Radon flux at the earth air interface. *Journal of Geoph. Res.*, **65**, No. 10, October.

W2. Wilkening, M. H., 1959, Daily and annual courses of natural atmospheric radioactivity. *Journal of Geoph. Res.*, **64**, pp. 521–526.

ВРЕМЕННЫЕ И ПРОСТРАНСТВЕННЫЕ ИЗМЕНЕНИЯ КОНЦЕНТРАЦИИ КОРОТКОЖИВУЩИХ ПРОДУКТОВ РАДИОАКТИВНОГО РАСПАДА РАДОНА В НИЖНЕЙ АТМОСФЕРЕ

Поскольку изучение естественной радиоактивности в нижней атмосфере может давать в будущем информацию о турбулентной диффузии и переносе воздушных масс, мы предприняли исследование состоящее из двух частей.

Во-первых мы изучили естественную радиоактивность атмосферы над плоскогорьем, во-вторых произвели измерения в различных местах земного шара для получения общего распределения радиоактивности в нижней атмосфере. Мы нашли, что радиоактивность над плоскогорьем намного больше, чем над океанами (100 и 6×10^{-12} Ci $м^{-3}$ соответственно) и в северном полушарии больше, чем в южном (6 и 15×10^{-12} Ci $м^{-3}$ соответственно).

Основываясь на этих данных мы считаем возможным с помощью естественной радиоактивности индивидуализировать массы воздуха и следить за вертикальной и горизонтальной диффузиями в них.