Aircraft Measurements of Rn-222, Aitken Nuclei and Small Ions up to 6 km

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(Manuscript received 11 June 1971, in revised form 24 September 1971)

ABSTRACT

Simultaneous measurements of the vertical distribution on Rn-222, Aitken nuclei and small ions have been carried out in southwest France, several hundred kilometers from the Atlantic Ocean, between ground level and 6000 m.

These measurements demonstrate the importance of radon as a tracer characterizing the continental nature of an air mass. Every one of the measurements shows an increase in the concentration of radon at altitudes in excess of 3000 m. The radon does not appear to have originated in the European continental land mass. The concentrations of Aitken nuclei, small ions and radon show a sudden variation at the upper limit of the planetary boundary layer which is due, at least in part, to the geographic location of the site at which the measurements were made.

With certain simplifying hypotheses, the mean size of atmospheric aerosols can be deduced from these measurements.

1. Introduction

The evolution of aerosols and pollutants of natural and of man-made origin within the atmosphere is a problem of considerable importance. In this respect the gas radon constitutes an interesting tracer. Radon is, in fact, a rare gas emanating essentially from large continental land masses. It does not react chemically with the environment and its lifetime is limited only by its radioactive half-life. The distribution of radon within an air mass thus provides an indication of its stability and of the time spent over a large land mass. It is therefore of interest to compare the vertical distribution of radon and of Aitken nuclei. The origin and evolution of the latter are, in fact, not well understood. They are mainly produced over continental areas and their origin may either be natural or man-made.

The concentration of small ions is a function of the ionization of the air (hence of the radon concentration) and of the concentration and mean size of the aerosols. Comparisons between the radon concentrations, the number of Aitken nuclei, and the concentration of small ions enable the mean size-distribution of the aerosols to be determined. As in the case of the Aitken nuclei, the small ions are thus indicators of particulate air pollution.

2. Basic concepts

a. Radon

Rn-222 is a radioactive gas (half-life 3.8 days) which is formed in the soil and released into the atmosphere in which it diffuses. Radon is produced mainly at the surface of large continental land masses; the release of radon from the oceans is very low (Broecker *et al.*, 1967; Broecker and Kaufman, 1970). As a consequence the concentration of radon in an air mass which has remained over the ocean for a period corresponding to several times the radon half-life is low.

Fig. 1 illustrates the evolution of radon concentration in an oceanic air mass displaced over a continental land mass. The curves were derived by the use of a model based on a numerical solution of the diffusion equation (Birot *et al.*, 1970). The diffusion profile used corresponds to the case where a stable zone exists at about 1000 m, i.e., at a level above that of the planetary boundary layer. It will thus be seen that the radon concentration reaches a limiting value after having travelled several thousand kilometers, mainly at higher altitude.

By way of comparison, Fig. 2 presents some profiles measured by various authors, over continental sites (curves 1, 2 and 4) or at several hundred kilometers from the coastline (curve 3).

b. Small ions

Small ions in the atmosphere are mainly produced by:

1) Radioactive elements in the atmosphere: radon, thoron and their decay daughters.

2) α, β and γ radiation from radioactive substances in the ground.

3) Cosmic radiation.

The respective influence of these three principal ionizing agencies is a function of altitude (Fig. 3).

The ionizing radiations emitted by the soil can only be detected within the first 100 m of altitude (Israel,

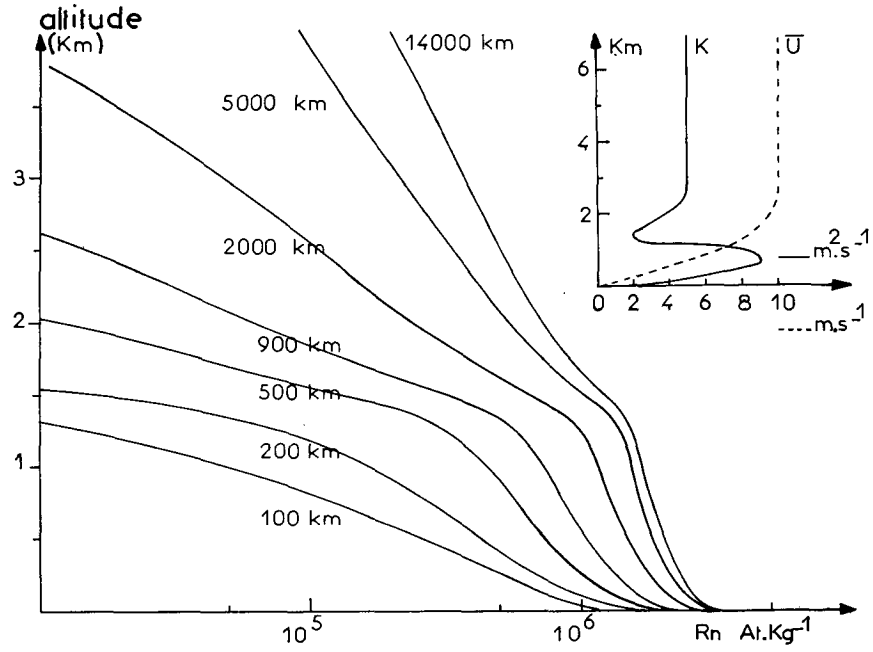

FIG. 1. The evolution of the vertical distribution of radon in an oceanic air mass displaced over a continental land mass for varying distances from the coastline.

1970) (curve 1 of Fig. 3); thus, its influence is primarily confined to the lower layers over a large land mass. Curve 2 of Fig. 3 represents the ionization corresponding to the mean distribution of radon obtained during the experiments to be described later. With altitude, cosmic radiation rapidly becomes the principal ionizing agent. Its intensity is a function of altitude and latitude of the point of measurement (curves 3, 4 and 5 of Fig. 3).

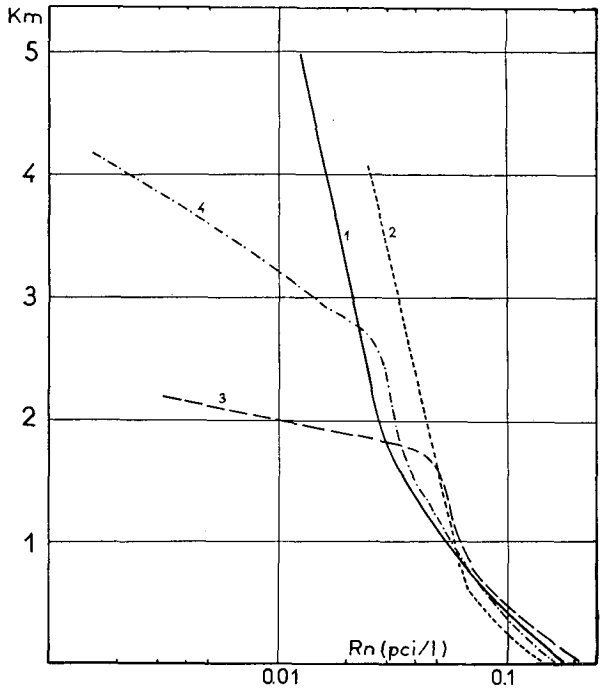

FIG. 2. Experimental profiles for radon: continental sites 1. (Kirichenko, 1962), 2. (Wilkening, 1970), 4. (Bradley and Pearson, 1970); coastal site 3. (Birot *et al.*, 1970).

Molecules of water become attached to the small ions when they are formed; their mobility is 2–0.5 $cm^2\ sec^{-1}\ V^{-1}$ (Huertas *et al.*, 1970). The mobility of the negative ions is larger than that of the positive ions, the mean ratio k^-/k^+ being 1.25. The small ions disappear either by recombination with those of opposite sign, or by attachment to electrically neutral or charged nuclei. The lifespan of these small ions is thus a function of particulate air pollution and of the intensity of ionization. Their lifetime can range from several seconds to several minutes. Thus, at ground level in the case of considerable particulate pollution, i.e., in the presence of 50,000 particles cm^{-3}, the lifetime of the small ions is of the order of 30 sec. It is of the order of 150 sec if the number of particles approximates 10,000. At higher altitudes, attachment to particles can often be neglected compared to recombination. The lifespan of the ions can then be taken to be of the order of 400 sec at 1500 m and 200 sec at 5000 m. The electrical conductivity of the air is mainly due to small ions.

c. Aitken nuclei

The origin and nature of Aitken nuclei remains ill-defined. It is believed that they may be formed by:

1) Condensation of substances in cooled air. In this way industrial smoke provides a large number

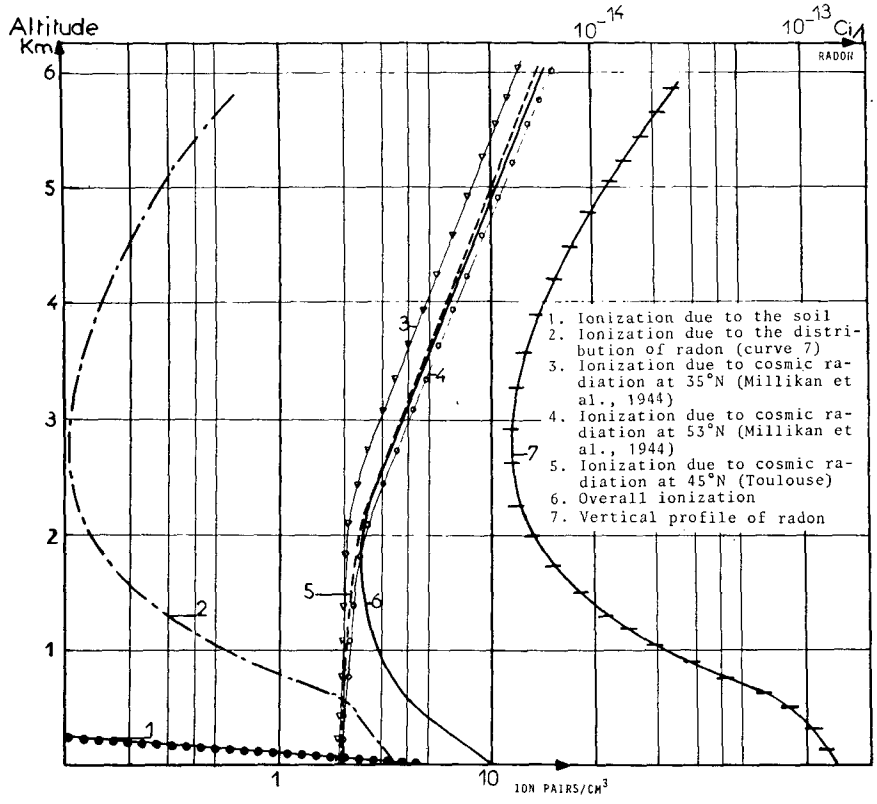

FIG. 3. The influence of various ionizing agencies.

of particles with dimensions in the size range of Aitken nuclei.

2) Photo-chemical reactions involving minor constituents in the atmosphere (for example, air pollutants). Aitken (1911) first demonstrated that the presence of sulphur dioxide in the air encourages the formation of nuclei under the influence of sunlight. This phenomenon has again been discussed by Verzar and Evans (1959) and, more recently, by Madelaine (1968) and Mohnen and Lodge (1969). However, the reactions which stimulate the formation of nuclei from photolytic action are not well understood. Other substances (nitrogen monoxide, ammonia and certain hydrocarbons) have been shown to generate nuclei. These substances are mainly found in urban areas and are particularly representative of pollution. In rural areas it is mainly the organic substances produced by vegetation which encourage the generation of nuclei. Went (1966) demonstrated that air containing α-pinene (a substance emitted by the resinous forests) can generate nuclei by photolytic action.

The mean size of Aitken nuclei is around 3×10^{-6} cm, (Junge *et al.*, 1961). The production of such nuclei by the oceans may be neglected (Ohta, 1951; Hess, 1951), since they are significantly larger than those of the Aitken variety (Blanchard, 1963). Thus, low concentrations of Aitken nuclei are to be found over oceans and coastal areas and, as in the case of radon, Aitken nuclei can be considered to be mainly of a continental origin.

Few measurements of the altitude profiles of Aitken nuclei exist at present: Wigand, 1919; Weickmann, 1955; and Imyanitov and Churabrina, 1967, at Leningrad. Reiter *et al.* (1970) have also measured the vertical profile of Aitken nuclei, up to 3000 m along the side of a mountain at Garmish. It should be noted that the authors of these measurements have not defined the origin and characteristics of the corresponding air masses.

3. Theoretical relation between the number of small ions, the intensity of ionization, and the number of Aitken nuclei in the atmosphere

a. Equation for the evolution of the small ions

The equations for the evolution of small ions can be expressed as (Bricard, 1965):

$$\partial n^+/\partial t = 0 = q - \alpha n^+ n^- - n^+(\beta_+{}^- N^- + \beta_+{}^0 N^0), \quad (1)$$

$$\partial n^-/\partial t = 0 = q - \alpha n^+ n^- - n^-(\beta_-{}^+ N^+ + \beta_-{}^0 N^0), \quad (2)$$

where q is the rate of ion pair formation; n^+ and n^- the concentrations of positive and negative small ions; N^+, N^- and N^0 the concentration of positive and negative large ions and neutral particles, respectively; α the

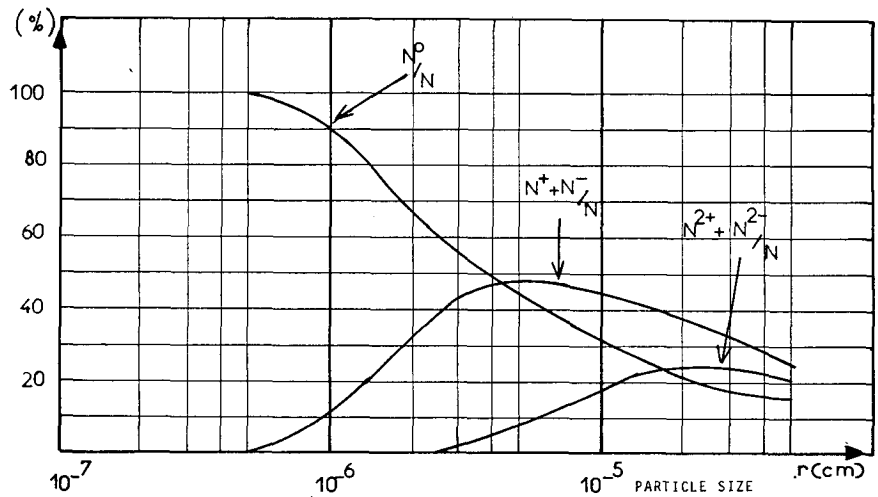

FIG. 4. Percentage of charged particles assuming Boltzmann equilibrium (Keefe *et al.*, 1959). See text for further information.

recombination coefficient for small ions; and β_i^p the attachment coefficient for small ions of polarity i to particles with p elementary charges. These coefficients have been determined by Bricard (1965) and Mohnen (1966). They are a function of particle size.

b. Variation of the ratio n^+/n^-

The solution of Eqs. (1) and (2) is

$$n^+/n^- = \beta_-^+/\beta_+^- (N^+/N^-)^{\frac{1}{2}} = K^-/K^+ (N^+/N^-)^{\frac{1}{2}} = 1.25(N^+/N^-)^{\frac{1}{2}}, \quad (3)$$

where K^+ and K^- are the mobility of positive and negative small ions, respectively.

Bricard *et al.* (1965) have determined (assuming a mean size of the aerosol of 3×10^{-6} cm) the variation of N^-/N^+ in terms of Q/N^2, the characteristic "factor of pollution" and of ionization. Effectively, $Q=q-\alpha n^+n^-$ and $N=N^0+N^+ +N^-$.

If $Q/N^2<10^{-7}$, as is generally the case in the lower atmosphere, $N^-/N^+=1$. Hence,

$$n^+/n^-=1.25.$$

If $Q/N^2>10^{-5}$, as is the case at higher altitude, $N^-/N^+=1.56$. Hence,

$$n^+/n^-=1.$$

c. Variation of the concentration of small ions as a function of number of particles and intensity of ionization

Within the lower layers where $n^+/n^-=1.25$ and $N^+=N^-$, Eq. (2) becomes

$$q-1.25\alpha(n^-)^2-n^-[(\beta_-^+/\beta_-^0)N^- + N^0]\beta_-^0=0. \quad (4)$$

The ratio β_-^+/β_-^0 is a function of the size of the particles (Bricard, 1965).

The ratio of the charged and neutral particles is also a function of the size of these particles. Assuming the Boltzmann equilibrium, Keefe *et al.* (1959) have determined the percentage of neutral nuclei and the percentage of those carrying 1, 2, . . . charges (Fig. 4).

Considering a mean radius of Aitken nuclei of 3×10^{-6} cm, the solution of (4) is

$$n^- = \frac{-\beta_-^0 N + [(\beta_-^0)^2N^2 + 5\alpha q]^{\frac{1}{2}}}{2.5\alpha}, \quad (5)$$

where N represents the total number of Aitken nuclei.

At higher altitudes, the attachment of the small ions to particles is insignificant relative to recombination. In this case

$$n^+=n^-=(q/\alpha)^{\frac{1}{2}}.$$

With some knowledge of the concentration of small ions, radon and Aitken nuclei, Eq. (4) allows the determination of β_-^0 corresponding to the Aitken nuclei present, and hence to their mean size. Thus, we have

$$\beta_-^0 = \frac{q-1.25(n^-)^2}{n^-[N^0+N^+(\beta_-^+/\beta_-^0)]}. \quad (6)$$

This equation is easily solved through successive approximations by taking any logical initial value for β_-^+/β_-^0.

4. Experimental apparatus

The measurements between ground level and 6000 m altitude were made from two aircraft flying simultaneously over the same area.

The first aircraft, a Pilatus Turbo-porter, was equipped with the radon measuring apparatus. The second aircraft, a Cessna TU.206, was equipped for the measurement of small ions, Aitken nuclei and meteorological parameters.

a. Meteorological parameters

Temperature, relative humidity, dew point and pressure were measured by means of a platinum wire, an activated carbon plate, a condensation hygrometer and a capsule type barograph, respectively. The outputs were recorded continuously.

b. Measurement of radon

The radon concentration was measured by means of two different instruments. The first (Birot *et al.*, 1970) is based on the collection of aerosols on a filter, making it possible to estimate the concentration of radon in the air at the moment of sampling.

A more precise measurement was obtained by the activated charcoal adsorption method. The sample air (average of 200 liters), after having been passed through a water and a CO_2 trap, passes through a cartridge of activated charcoal (30 gm, cooled by dry ice), in which the radon is adsorbed. The rate of flow is of the order of 30 liters min^{-1}. The total weight of the installation, which comprises a maximum of 10 sampling cartridges, is 40 kg and the electrical consumption 140 W. The determination of the quantity of radon contained within the cartridges is carried out in the laboratory. After having heated the charcoal to 400C, the radon is extracted and transferred, with the aid of a carrier flow of helium, into a scintillating balloon flask having a capacity of 500 cm^3. This latter is placed in front of the window of a photomultiplier 3 hr after filling. The mean sensitivity is 5 counts per minute (cpm) $(pCi)^{-1}$ of radon (Delos, 1970). The passage of 200 liters of air through the cartridge enables the measurement of 10^{-15} Ci $liter^{-1}$ to within an accuracy of 15%.

c. Measurement of Aitken nuclei

The nuclei are measured with a self-contained portable counter (Fig. 5) constructed in our laboratory and based on the principle of the Aitken counter (Cabrol, 1970). The sample air is introduced into a cylindrical chamber 30 cm in length and 3 cm in diameter. A water reservoir attached to the cylinder permits the air to become saturated with water vapor. The air is compressed slowly and then rapidly expanded. Thus, the Aitken nuclei present in the chamber play the role of condensation nuclei and the attenuation of a beam of light by the droplets formed around the nuclei is measured. The apparatus, calibrated theoretically and by comparison with a General Electric counter, enables measurements to be made at intervals of 1 min.

The particles which have a radius $>7\times10^{-8}$ cm are measured. The lowest sensitivity is 300 particles cm^{-3}.

The accuracy of measurement is a function of the nuclei concentration: 3% for 20,000 particles cm^{-3} and 25% for 1000 particles cm^{-3}.

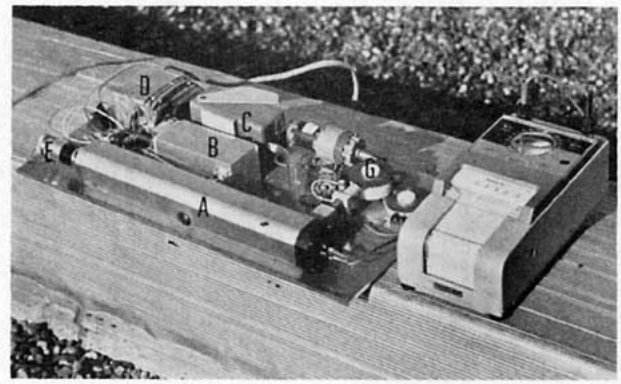

FIG. 5. Apparatus for the measurement of Aitken nuclei: A, measuring chamber; B, compression chamber; C, pump; D, motor; E, optical system; F, programmer

d. Measurement of small ions

The positive and negative ion concentrations were measured by means of a Gerdien condenser operating in the saturation range as defined by its current-voltage. The ion precipitators, mounted below the wing of the aircraft, are identical in mechanical construction for both positive and negative ions. The airstream through the ion precipitators is provided by the forward speed of the aircraft. The electrical field between the electrodes of the condenser is controlled such that the ions of mobility greater than 0.4 cm^2 sec^{-1} V^{-1} (at ground level) are collected by the ion precipitators. The ion current is measured at the axial electrode by means of an electrometer whose potential is isolated from the chassis, thus enabling a high potential to be applied to the measuring electrode to better define the electrical field, particularly at the entrance. With increasing altitude, the high voltage is continuously adjusted to maintain the limit of mobility for ion collection at the ground level value of 0.4 cm^2 sec^{-1} V^{-1}. The forward speed of the aircraft during sampling is generally in the neighborhood of 46 m sec^{-1}, which corresponds to a Reynolds number of the order of 8000 in the Gerdien condenser. The accuracy is 5%.

5. Experimental results

The simultaneous concentration measurements of radon, Aitken nuclei and small ions between ground level and 6000 m were made on 5, 6, 10 and 11 July 1970 in the vicinity of Toulouse at a distance of 250 km from the coast. The meterological conditions were as follows:

1) At ground level (<1000 m). An anticyclone, centered over Europe, gave northeasterly winds on the 5th of July and a fairly strong southeasterly wind on the 6th. On 10 July, southwest France was on the extreme edge of an anticyclone centered over the Atlantic, with light winds having a northerly component. On 11 July a front with irregular winds veering between

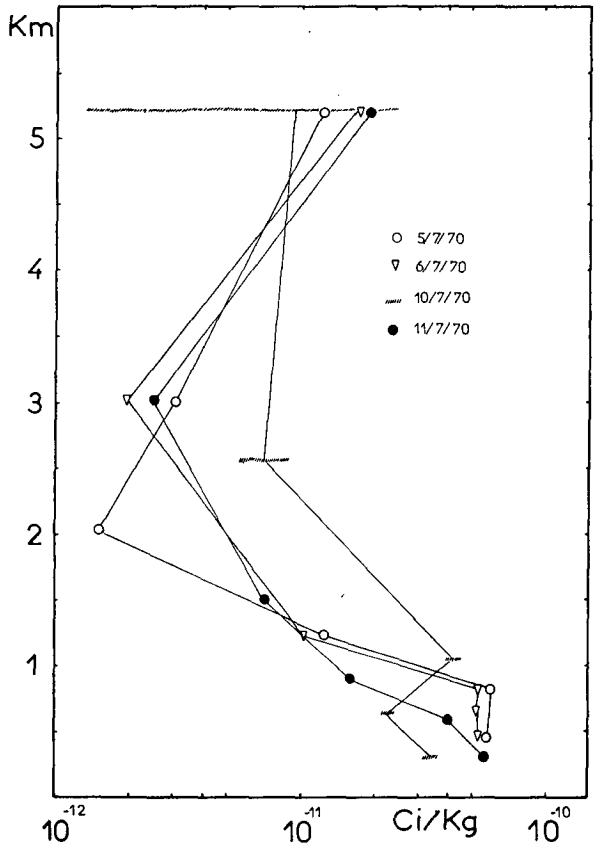

FIG. 6. Radon profiles. (The one for 10 July was obtained with the "filter" apparatus.)

northwest and northeast approached the southern part of France.

2) At intermediate levels (1000–2000 m). An anticyclone, centered over the Gulf of Gascogne on 5 July gave a light airflow from east to northeast. The movement of the high pressure area toward North Africa on the 6 July resulted in an airflow from the west. On 10 and 11 July winds aloft were west-northwesterly.

3) Above 3000 m. Throughout the period of our measurements, winds aloft were west-northwesterly.

a. Discussion of the measurements of radon

In the lower layer, below 1500 m, the radon profiles change with the sampling day (Fig. 6). Thus, on 5 and 6 July the concentration is practically constant (~50 pCi kg^{-1}) up to 1000 m, with a rapid decrease above this altitude. On both days a stable layer was found at approximately 1000–1200 m, which prevented the radon from diffusing higher. The effect of the accumulation below this stable layer is accentuated by turbulence of mechanical origin, particularly on 6 July when the southeast wind reached a speed of 12 m sec^{-1}. (This turbulent state was confirmed by the in-flight recorded parameters.)

The total quantity of radon content in the layer from 0 to 1000 m is larger on the 5th and 6th of July than on the 10th and 11th. On both the 5th and 6th, the air in the lower layer had remained longer over the continent (with weak northeast and southeast trajectories, respectively) than on the 10th and 11th when the air (with a westerly trajectory) came more directly from the ocean. In particular, the profile obtained on the 11th is similar to that obtained with the half-plane model (Fig. 1) for a distance of several hundred kilometers over the continent. The effect of the temperature inversion at 1000 m is less noticeable, the percentage of radon being lower.

In all flights an increase in the radon concentration was observed above 3000 m. Typically a concentration of 10–20 pCi kg^{-1} was obtained at 5000 m. We have previously observed a similar variation of radon with altitude (Birot *et al.*, 1970). The altitude profile for the radon concentration as presented in Fig. 6 can be understood on the basis of Machta's (1961) interpretation that he derived from radon profiles obtained on the California coast with air masses arriving from the continent of Asia, i.e., that the greater concentrations at higher altitude could be due to differences in the transit time over the ocean. This phenomenon of radon transport over an ocean by advection was also reported by Prospero and Carlson (1970), between Dakar and Barbados.

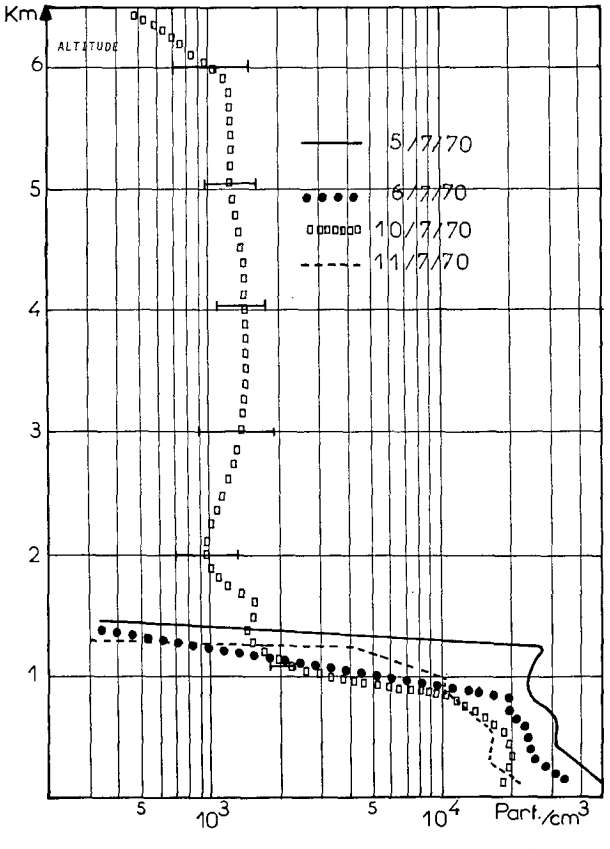

FIG. 7. Measured altitude profile of Aitken nuclei.

With respect to our measurements, the radon content in low layers proceeds from the European continent. The radon increase above 3000 m probably comes from an advection phenomenon; the meteorological measurements over the North Atlantic Ocean are not very numerous and it is difficult to determine exactly the isentropic air mass trajectories. Nevertheless, considering the normal western flux above 3000 m, we can probably say that the radon is coming from the American continent.

b. Discussion of the measurements of Aitken nuclei and of small ions

A rapid diminution of the concentration of Aitken nuclei was observed daily when passing through the boundary layer (Fig. 7). As in the case of radon, the total quantity of nuclei contained in the layer from 0 to 1200 m is greater for the 5th and 6th than for the 10th and 11th of July, a fact that appears to confirm the continental origin of these particles. The weak nuclei concentration (lower than the sensitivity of apparatus) above 1200 m might be explained by the oceanic trajectory of the air masses. Conversely, on 10 July a profile practically constant and equal to 1000 particles cm^{-3} was found between 1200 and 6000 m. Considering the 200-km air mass travel over the continent and the stability of the atmosphere above 3000 m, it is unlikely that these nuclei were of European continental origin.

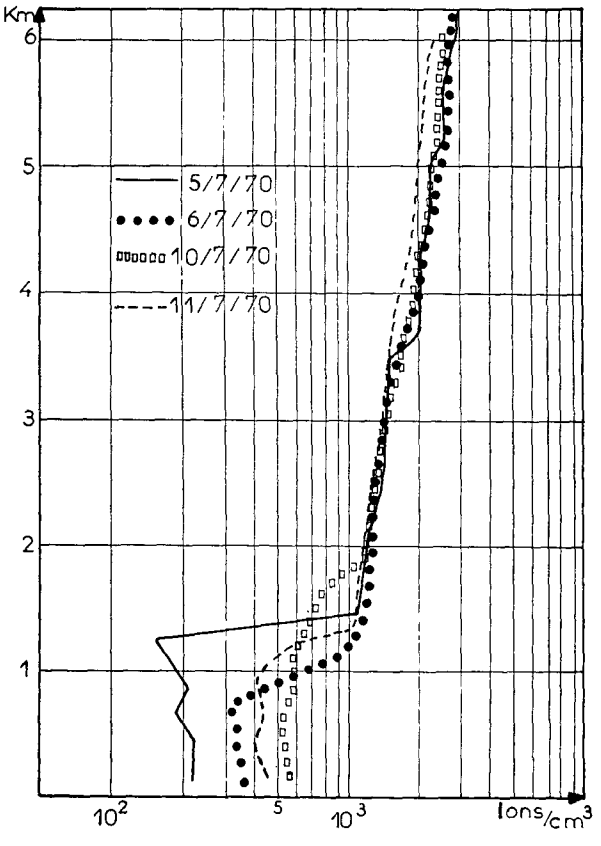

FIG. 8. Measured altitude profile of small ions.

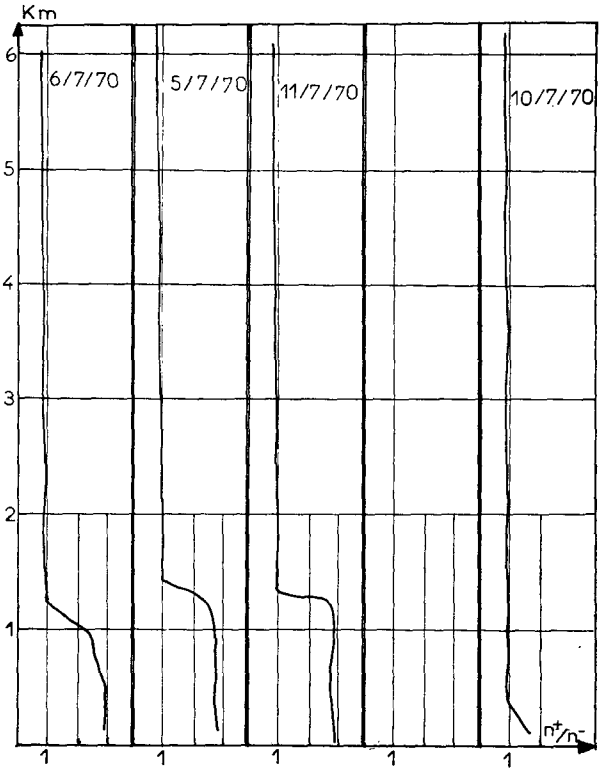

FIG. 9. Variation of the ratio n^+/n^- as function of altitude.

In the case of the small ions (Fig. 8), their increase at altitudes >1200 m corresponds with the disappearance of the nuclei. The same curves were derived by Sagalyn and Faucher (1954) who measured the conductivity of the air over New England. These authors attributed the sudden rise in conductivity to the different conditions of diffusivity existing on both sides of the upper level of the boundary layer. We believe that the rapid variations which we obtained are due both to the stable layer overlying the boundary layer in each of our experiments, and to the fact that the upper air was of oceanic origin having a low concentration of nuclei.

The ratio n^+/n^- is presented in Fig. 9. On 5, 6 and 11 July we obtained a mean value of 1.2 within and 1.0 above the boundary layer. This gives close agreement with the theoretical values which were to be expected.

On the 10th, however, n^+/n^- is practically equal to unity irrespective of altitude. This result is difficult to interpret. It could be due to differences in aerosol size. We noted the presence of a cloudy zone at approximately 1000 m on that day.

Using relations (4) and (6) and the experimental concentration profiles of small ions, Aitken nuclei and radon, we have determined the value of the mean

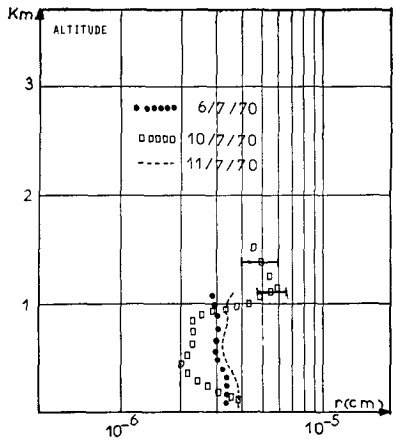

FIG. 10. Variation in the calculated mean radii of aerosols as a function of altitude.

radius r of the aerosols (Fig. 10). The accuracy is

$$\frac{\Delta r}{r} \approx \frac{\Delta \beta_{-}^{0}}{\beta_{-}^{0}} \approx \frac{\Delta n^{-}}{n^{-}} + \frac{\Delta N}{N}.$$

The mean size that we obtained was similar for the 6th and 11th of July, being practically constant with altitude and corresponding to a mean radius of the order of 3.5×10^{-6} cm. The profile obtained on the 10th is different. The mean radius at ground level is the same as that observed for the other days and decreases up to 400 m. From 400 to 900 m the mean size of the nuclei is of the order of 2×10^{-6} cm. It would thus appear that there exists a zone in which the nuclei have smaller dimensions. Above this level, there is a sudden increase in the mean radius from 2×10^{-6} to 6×10^{-6} cm. This increase corresponds to the cloudy zone whose presence was indicated earlier.

6. Conclusions

During each sampling we observed an increase in the concentration of radon above 3000 m. This concentration reaches values of 10–20 pCi kg^{-1} at about 5000 m. The radon, which could not have originated over the European land mass, had probably been carried over the Atlantic by advection from the American continent. Radon thus constitutes an interesting tracer for the transport of pollutants on a synoptic scale. In our next series of experiments we expect to measure the concentrations of certain pollutants, such as sulphur dioxide, sulphates and lead, as well as Aitken nuclei, with greater sensitivity. In effect, with the exception of the samplings made on 10 July, the concentrations of Aitken nuclei aloft were lower than the sensitivity of the apparatus employed.

The concentrations of Aitken nuclei, of small ions, and of radon present a sudden variation at the upper level of the planetary boundary layer. This provides good evidence for the continental origin of Aitken nuclei. These variations at the level of the boundary layer must often occur at other sites. We believe, however, that they are accentuated, as far as our results are concerned, by the geographical location of France which, above the boundary layer, is supplied with air originating from the Atlantic.

With certain simplifying hypotheses, the mean dimension of atmospheric aerosols can be deduced from these measurements. It is hoped that, in the near future, we shall also be able to measure the number of large ions, within the planetary boundary layer, which will help us to reduce the number of assumptions. In addition, a diffusion battery placed before the nucleus counter will provide us with another method of determining the mean value of the size of the aerosol.

Acknowledgments. We wish to express our thanks to Prof. D. Blanc, Director, Centre de Physique Atomique et Nucléaire, for the great interest that he has shown in this study. The in-flight measurements were carried out with the collaboration of Messrs. Delos (radon), Cabrol (Aitken nuclei), Boyer and Domergue. We are grateful, too, to M. Druilhet for the fruitful discussions we shared in respect to the interpretations of our measurements. Throughout the period of these measurements we enjoyed the benefit of the collaboration of the Meteorological Station of Toulouse-Blagnac and our thanks are due to M. Cazale, Head of Service, who facilitated our task. Finally our thanks are due to Messrs. Jalu and Gland of SMM of the Service Météorologie Nationale.

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