Vertical Exchange Measurements in the Lower Troposphere Using ThB (Pb-212) and Radon (Rn-222)

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ABSTRACT

Various methods of determining vertical eddy diffusion coefficients in the planetary boundary layer from two natural radioactive tracers, radon (Rn-222) and ThB (Pb-212), are presented and discussed. In all cases, it is more pertinent to measure vertical exchanges in the lower atmosphere using ThB (10.6 hr half-life) than radon (3.8 days half-life).

About 30 vertical profiles of Rn and ThB in the lower troposphere (100–3000 m) are determined over a 12-month period. Vertical eddy coefficient values are calculated according to the various methods discussed. The variation range, between 10^{-1} and several 100 m^2 sec^{-1}, is large and closely related to the vertical temperature gradient variation.

Low values (10^{-2} to 10^{-1} m^2 sec^{-1}) of the exchange coefficients determined in the inversion layer which frequently exists at the top of the planetary boundary layer show that vertical turbulent fluxes toward the free atmosphere are relatively small.

1. Introduction

Studies now being conducted on medium- and long-term weather forecasting are based on the use of numerical models. In order to simplify such models, it is possible to disregard the presence of the planetary boundary layer, the site of complex phenomena difficult to introduce into a global-scale numerical model. Vertical fluxes between this layer and the free atmosphere must, however, be introduced as a boundary condition; it is therefore important to determine the exchange coefficients near the ground. Moreover, since the intensity of vertical exchanges is a decisive factor in the efficiency of effluent dispersion in the atmosphere, measurement of these exchanges, especially in the planetary boundary layer, can be applied to problems of atmospheric pollution.

The turbulent vertical flux of a scalar quantity can be written as

$$F=\overline{\rho w' a'},$$

where ρ is air density, $w=\bar{w}+w'$ (the vertical wind component) and $a=\bar{a}+a'$ (the concentration per mass unit of diffusion quantity).

Certain authors have made direct measurements of w' and a' in the surface layer (0–100 m). Such measurements, however, require sensitive and delicate equipment which is difficult to operate at ground level, let alone from aircraft. Turbulent fluxes can also be obtained from mean values using eddy diffusion coefficients ("K" theory). The K theory can be used if the scale of turbulent motion is small compared to the scale of measurement. The turbulent flux in terms of mean values can be written as

$$F=-\rho K\frac{\partial \bar{a}}{\partial z}.$$

The coefficient K is independent of the nature of the diffusing quantity and depends only upon atmospheric thermodynamic characteristics, but the quantity considered must be conservative and "passive" in regard to turbulence. Since heat and momentum can be related to the motion of tracers, it is possible to define a transfer coefficient for heat (K_θ) and for momentum (K_m) (Obukhov, 1971). For several years natural radioactive tracers have been used to measure the vertical exchange of matter. These studies are generally based upon the use of radon, which, because of its 3.8 day half-life, is influenced by large-scale motions (Shopauskas, 1964; Assaf, 1969, 1971; Birot *et al.*, 1970; Huss *et al.*, 1970; Nazarov *et al.*, 1970). Another natural tracer, ThB (Pb-212), whose half-life is 10.6 hr, seems better suited to measuring exchange coefficients on an aerological scale. Assaf and Biscaye (1972) were first to apply ThB to the case of exchanges in an urban boundary layer; Guedalia *et al.* (1973) applied it to the case of exchanges in the planetary boundary layer. This tracer had already been used to measure vertical exchanges in the surface layer (0–100 m) (Druilhet, 1966; Druilhet and Fontan, 1972). Assaf (1969) and Beran and Assaf (1970) showed how careful one should be when using K theory with radioactive tracers. ThB has the advantage of allowing the integration of all microscale motions without being sensitive to synoptic-scale motions.

We present first the methods which lead to a determination of vertical diffusivity in the planetary boundary layer with ThB and radon and a critical discussion. Several series of aircraft measurements which produced approximately 30 ThB and radon vertical profiles were carried out between October 1971 and October 1972. These measurements were taken in the Toulouse area (southwest France) about 200 km from the Atlantic coast. The variation of the exchange coefficients in the planetary boundary layer has been studied, and the influence of thermal gradient is shown.

2. Determination of K_z

The equation of diffusion in the case of a radioactive tracer can be expressed as

$$\frac{\partial \bar{C}}{\partial t}+\bar{u}\frac{\partial \bar{C}}{\partial x}=-\frac{1}{\rho}\frac{\partial}{\partial z}\left(\rho K_z\frac{\partial \bar{C}}{\partial z}\right)+\frac{1}{\rho}\frac{\partial}{\partial y}\left(\rho K_y\frac{\partial \bar{C}}{\partial y}\right)-\lambda\bar{C}, \quad (1)$$

where $\bar{C}$ is the mean concentration of tracer (atoms per kilogram of air), $\bar{u}$ the mean wind velocity in the x direction (m sec^{-1}), K_z, K_y the turbulent diffusion coefficients (m^2 sec^{-1}), and λ the decay constant of the tracer (sec^{-1}). Diffusion along the x axis, compared to the transport by advection, was assumed to be negligible; the vertical wind component $\bar{w}$ was taken to be zero.

In all cases the exchange coefficient at a level z_1 can be inferred from Fick's first law

$$K(z_1)=-\frac{\Phi(z_1)}{\left(\rho\dfrac{\partial \bar{C}}{\partial z}\right)_{z=z_1}}, \quad (2)$$

where $\Phi(z_1)$ is the vertical flux of tracer at level z_1. Therefore, the determination of $K(z)$ implies a correct calculation of the flux and of the concentration gradient. Various methods which permit calculation of the flux $\Phi(z)$ will be discussed later. In order to illustrate the validity of various assumptions concerning the calculation of exchange coefficients, a number of numerical experiments were carried out with a diffusion model. Eq. (1) is solved numerically by an implicit method, a description of which can be found in Birot *et al.* (1970).

Thus, from the theoretical K_z profile, it is possible to calculate the vertical distribution of the tracer. Then, the K_z profile is re-calculated using one of our methods, to determine the accuracy of the K_z calculation.

a. Steady-state conditions

1) Infinite homogeneous source

This is the most frequently used hypothesis; in this case, where $\partial\bar{C}/\partial x=\partial\bar{C}/\partial y=0$, Eq. (1) becomes

$$\frac{\partial}{\partial z}\left(\rho K\frac{\partial \bar{C}}{\partial z}\right)-\lambda\rho\bar{C}=0, \quad (3)$$

and the vertical flux is

$$\Phi(z_1)=\int_{z_1}^{\infty}\lambda\rho\bar{C}dz=\int_{z_1}^{z_M}\lambda\rho\bar{C}dz+\underbrace{\int_{z_M}^{\infty}\lambda\rho\bar{C}dz}_{\mathrm{R}}, \quad (4)$$

where z_M is the highest level of measurement. An approximate value of the second integral (R) can be obtained from extrapolating the upper concentration values and from assuming that the exchange coefficient is constant above the z_M level; i.e.,

$$\mathrm{R}=\frac{\lambda}{\alpha}\bar{C}(z_M), \quad (5)$$

where $\alpha=(\lambda/K_A)^{\frac{1}{2}}$, where K_A is the coefficient for $z>z_M$. This method makes it possible to determine the concentration with good accuracy, since the value of R is low compared to the total flux, i.e., the concentrations decrease rapidly in the upper layers. The method is much more effective, however, with ThB than with Rn because of the longer half-life of the former.

In some cases, the vertical flux cannot be inferred from (4) because there is little decrease of concentration with altitude. In these cases, it is possible to approximate values of the exchange coefficients by dividing the atmosphere into layers and to assume that the coefficient K is constant in each of them. In this case we have

$$K_z=\frac{\lambda(\Delta z)^2}{(\log C_1/C_2)^2}, \quad (6)$$

where Δz is the thickness of the layer, and C_1, C_2 the concentrations at the highest and lowest levels of the layer. This approximation, used by several authors in the case of radon (Shopauskas, 1964; Nguyen Ba Cuong, 1968; Reiter, 1969) can lead to an error (under- or over-estimation) which depends upon the values of the exchange coefficients above the layer considered and, for that reason, was not used here.

2) Non-homogeneous source: Ocean-continent discontinuity

The hypothesis of an infinite source is an approximation which has been used by numerous authors but which does not apply to all cases.

The emanation rate of Rn and of Tn from the soil depends upon both the soil's geological composition and its vegetation. Several authors have measured the emanation rate of radon (Pearson, 1965; Malakhov *et al.*, 1966; Kirichenko, 1970; Druilhet and Fontan, 1972) and of thoron (Guedalia, 1967; Laurent, 1969; Crozier, 1969; Guedalia *et al.*, 1970; Israel and Horbert, 1970; Druilhet *et al.*, 1972). The mean emanation rate from soil ranges from 10^{-12} to 10^{-13} Ci m^{-2} sec^{-1} for Rn and from 10^{-10} to 10^{-11} Ci m^{-2} sec^{-1} for Tn.

The emanation rate of radon from the oceans can be calculated from the vertical distribution of Rn-222 and Ra-226 in sea water; the deficit of Rn near the surface corresponds to the quantity emanating into the atmosphere. Chi Trach Hoang and Servant (1972) estimated this emanation rate from profiles measured by Mathieu (1970) and by Ku *et al.* (1970) in the Antarctic Ocean and by Broecker *et al.* (1967) and Broecker and Peng (1971) in the Atlantic and Pacific Oceans. The results obtained were 6×10^{-16} (Atlantic Ocean), 2.5×10^{-15} (Pacific Ocean) and 7×10^{-15} Ci m^{-2} sec^{-1} (Antarctic Ocean). Thus, in the Northern Hemisphere, radon emanation from the oceans is less than 1% of that from the continents. In the case of ThB, any oceanic contribution can be ignored. This means that there exists an important discontinuity of the source of radon and ThB at ocean-continent separations which leads to strong horizontal concentration gradients.

Assuming that the mean wind direction is perpendicular to the discontinuity (x axis) and that the source is infinite in the y direction, i.e., $\partial\bar{C}/\partial y=0$, the diffusion equation can be written

$$\bar{u}\frac{\partial\bar{C}}{\partial x}=-\frac{1}{\rho}\frac{\partial}{\partial z}\left(\rho K_z\frac{\partial\bar{C}}{\partial z}\right)-\lambda\bar{C}, \tag{7}$$

from which the vertical flux at level z_1 can be inferred:

$$\Phi(z_1)=\int_{z_1}^{\infty}\bar{u}\rho\frac{\partial\bar{C}}{\partial x}dz+\int_{z_1}^{\infty}\lambda\rho\bar{C}dz. \tag{8}$$

When the horizontal gradient $\partial C/\partial x$ tends toward zero, the vertical flux tends toward the value corresponding to the case of an infinite source [Eq. (4)].

Numerical Example

Eq. (7) has been solved numerically by assuming zero concentrations of Rn and ThB above the discontinuity. The diffusivity profile used corresponds to a mixing layer under 900 m over which lies a stable layer 200 m thick; the wind profile increases with altitude (Fig. 1). Fig. 2 shows the changes of vertical profiles and Fig. 3 gives the change of the vertical flux of Rn and ThB with distance from the coast The experimental results confirm the great variation of tracers concentration far from the coastline:

The changes of Rn and ThB concentrations measured during a flight between the Atlantic coast and the Toulouse area on 28 May 1972 are shown for comparison (Fig. 4). Three profiles were measured downwind: above the coast, at 100 km, and at 220 km inland.

b. Determining exchange coefficients

The horizontal concentration gradient does not allow Eq. (4) to be applied to calculating the vertical flux (and from that, the exchange coefficient). However, the vertical flux can be computed numerically using experimental profiles of ThB or Rn, separated by a distance Δx down the mean wind (Birot, 1971; Guedalia *et al.*, 1971).

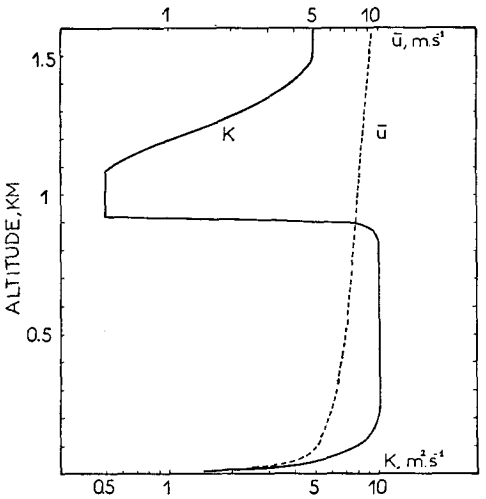

FIG. 1. Diffusivity (K) and wind (u) profiles used to calculate the Rn and ThB concentrations given in Fig. 2.

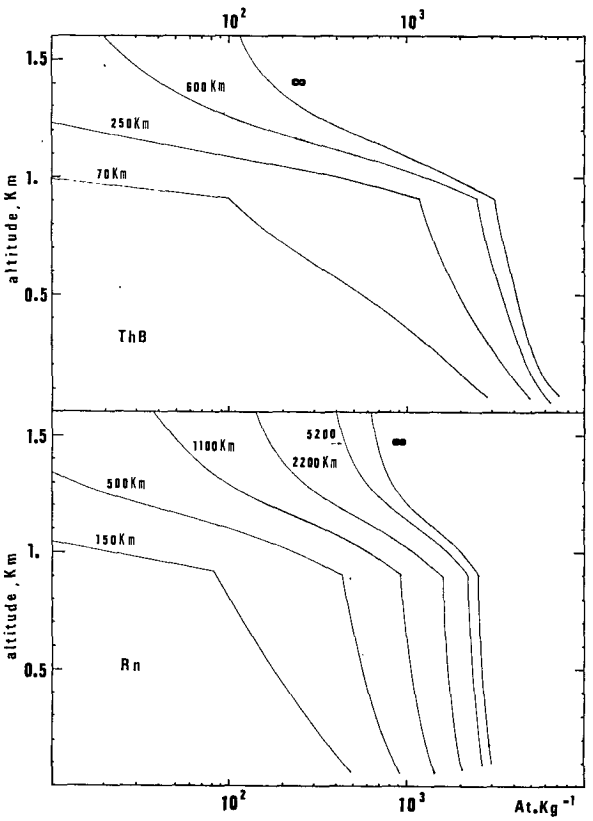

FIG. 2. Evolution of ThB and Rn vertical profiles for various distances from the discontinuity, the ∞ profile corresponding to an infinite source.

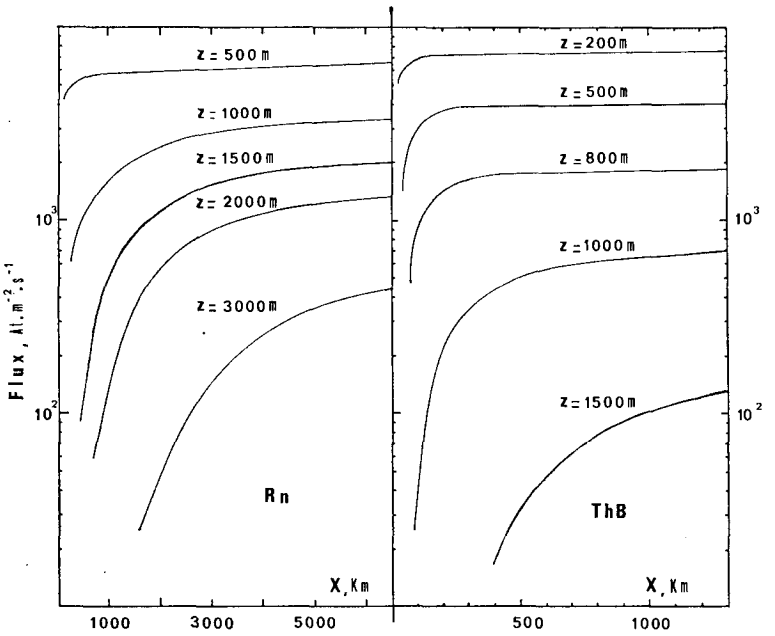

FIG. 3. Evolution of ThB and Rn vertical fluxes as a function of the distance from the discontinuity.

If the concentrations of the tracer above the discontinuity are negligible, the vertical flux can be calculated by using a single experimental profile measured at a distance Δx from the coast. The calculation is more accurate for ThB than for radon because of their respective half-lives.

c. Numerical experiments

Vertical diffusivity coefficients corresponding to profiles given in Fig. 2 have been calculated, taking into account the discontinuity of the source (for the particular case of zero concentrations above the coast). Fig. 5 shows the diffusivity profiles obtained by this

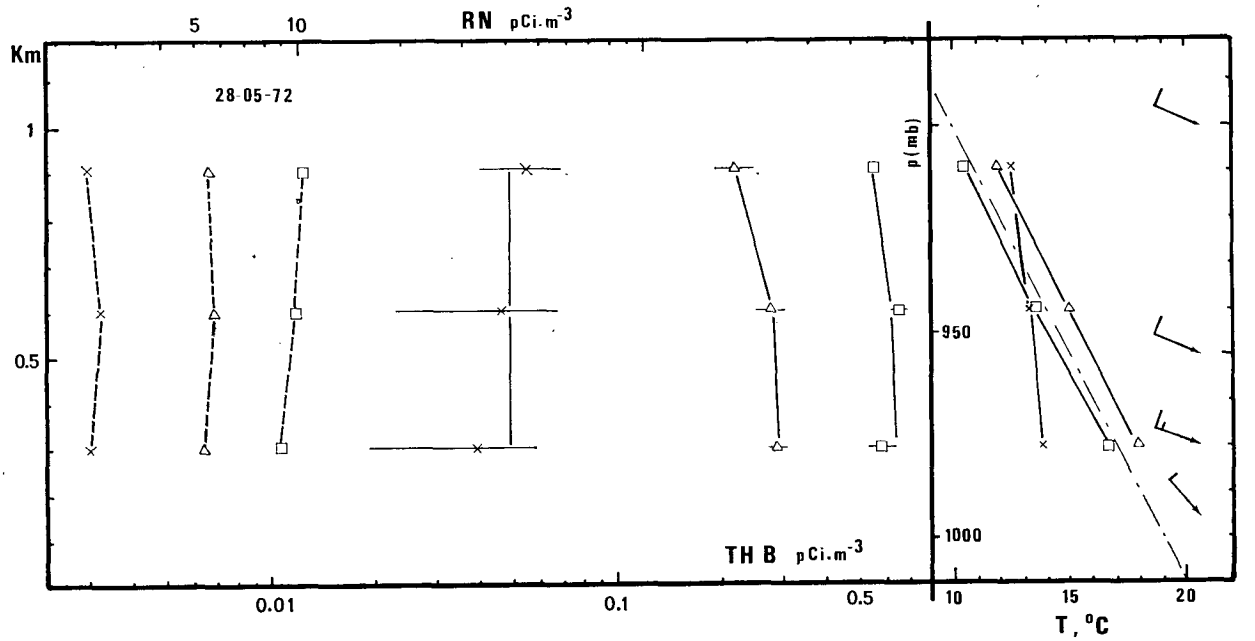

FIG. 4. Radon (dashed lines) and ThB (solid line) profiles obtained during the 28 May 1972 flight: □, 220 km from the coast; △, 100 km from the coast; ×, 5 km from the coast.

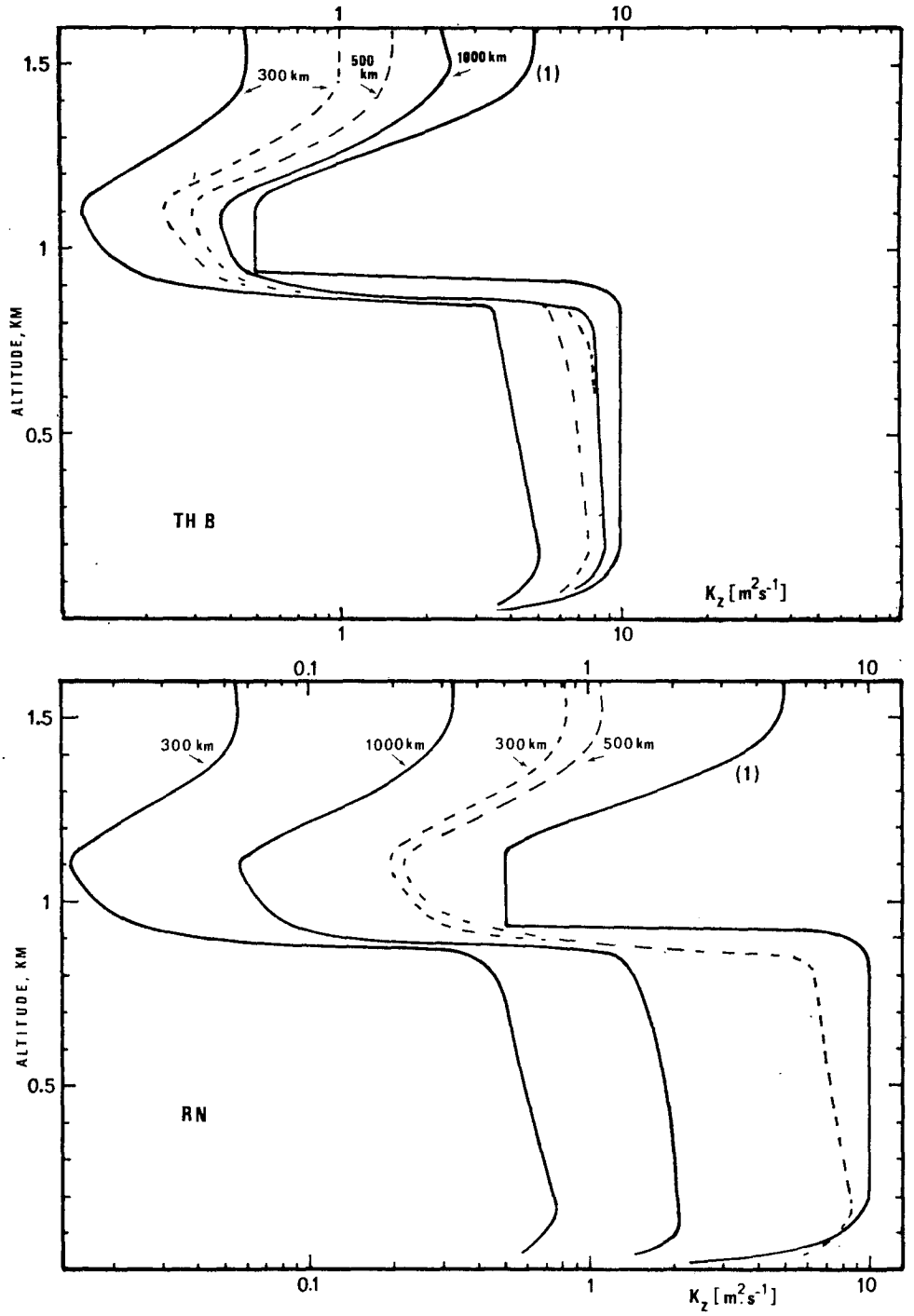

FIG. 5. K_z profiles derived from the Rn and ThB profiles given in Fig. 2: solid lines, infinite source hypothesis [Eq. (4)]; dashed lines, two-profile method [Eq. (8)]. The profile (1) is the initial K_z profile.

method (dotted lines) and those obtained in the case of an infinite source (solid line). It can be noted that:

1) Assuming the source to be an infinite plane results in underestimating the exchange coefficients, even far from the discontinuity. This error grows larger as $\partial\bar{C}/\partial x$ becomes more important, a situation likely to occur in the case of radon.

2) When the two-profile method (measured plus assumed zero profile above the discontinuity) is applied, the underestimation of the exchange coefficients is less than in the case of the infinite source hypothesis. This

method can be better applied in the case of ThB, for the zero-concentration hypothesis for air masses coming from the ocean is more plausible than in the case of Rn. This was the method we used to determine the vertical exchange coefficients from our aircraft measurements.

d. Cases of unmeasurable vertical gradients

Various methods for determining the tracer vertical flux have been presented. But in certain cases it is not experimentally possible to measure the gradient $\partial\bar{C}/\partial z$. Two such cases can be considered:

1) It has been frequently found that the $\partial\bar{C}/\partial z$ gradient cannot be measured experimentally for an unstable layer which features strong vertical mixing and which therefore maintains the measured concentration of the tracer constant. This condition likewise prevents the determination of the value K_z, yet it is possible to calculate a lower limit of the exchange coefficient. This minimum value is obtained by considering the strongest possible gradient, taking into account experimental errors for the measurements.

This type of calculation has several consequences:

1. Neither the real value of the exchange coefficient nor its form of variation in the mixing layer is known; its lower limit only can be determined.

2. If the experimental error for each measurement is constant, the gradient $\partial\bar{C}/\partial z$ obtained by this method will become smaller as the atmospheric layer becomes thicker. This means that the calculated lower limit of K_z will increase as the thickness of the considered layer increases.

It is possible to calculate the limiting value of K_z that can be measured as a function of the thickness of the layer. This calculation for ThB (Fig. 6) was conducted under four different diffusivity conditions above the mixing layer. An experimental error of 5% was allowed for each measurement. For example, for a layer $H=1$ km thick, it is not possible to determine values of K_z exceeding 90 m^2 sec^{-1} (with total reflection above the layer) or 300 m^2 sec^{-1} (with a value above the layer of $K_z=30$).

2) In the case of strong stabilities, the concentration gradient is always important. In certain situations, the concentrations drop to less than the sensitivity of the apparatus, thus preventing the measurement of the gradient $\partial\bar{C}/\partial z$. Under these circumstances, it is possible to determine a limiting value for K_z greater than the true value by replacing the unmeasurable concentration with the detection limit.

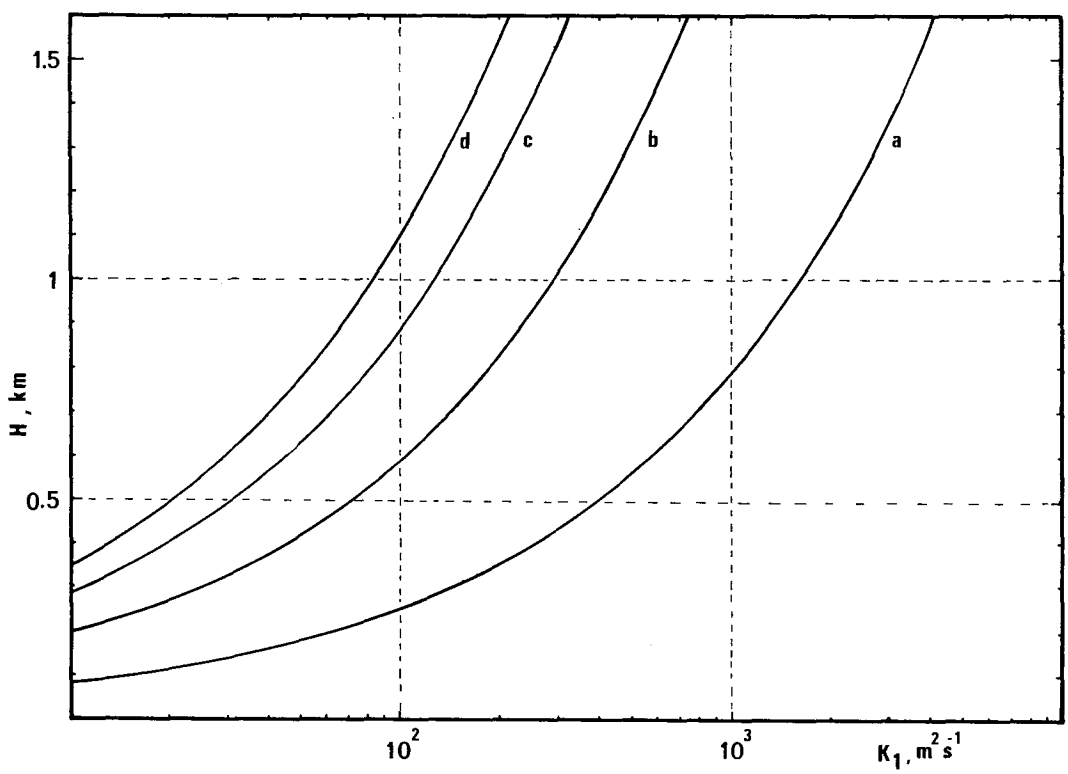

FIG. 6. Limiting value of K_z which can be determined using ThB as a function of the layer thickness (experimental error for each measurement, 5%): K_1, coefficient in the layer having a thickness H; K_2, coefficient above.
a) $K_2=K_1$ c) $K_2=0.01K_1$
b) $K_2=0.1K_1$ d) $K_2=0$

TABLE 1. Relative efficiency and background of the installations.

	Type of installation						
	α-β coincidence				α-scintillation		
	1*	2	3	4	5	6	7
Relative counting efficiency (%)	100	85±2	103±2	78±2	550±10	660±10	560±10
Background (counts per hour)	9	8	4	1.3	140	150	155

* Absolute efficiency is 5.2%.

e. Conclusion

The eddy coefficient K_z was derived from the relation

$$K(z_1) = -\frac{\Phi(z_1)}{\left(\rho\frac{\partial \bar{C}}{\partial z}\right)_{z=z_1}}.$$

Using ThB makes it possible to determine the flux by applying the infinite-source hypothesis [Eq. (4)] or by taking into account the ocean-continent discontinuity which produces horizontal concentration gradients [Eq. (8)]. Since the influence of this discontinuity on ThB calculations is negligible some hundred kilometers from the coast, Eq. (4) is regularly used for determining the flux. But in the case of an oceanic air mass covering a short distance over the continent, radon calculation of $\Phi(z)$ is only possible if Eq. (8) is applied. It has been assumed up to this point that no scavenging occurs. Actually, for this reason, the use of ThB is limited to clear weather or slightly cloudy situations. In the case of overcast skies, the vertical profile can be perturbed by scavenging phenomena, and profiles must be interpreted cautiously.

3. Experimental technique

a. ThB measurement

Since ThB (Pb-212) is a β-emitter, its concentration is easily derived from that of its daughter, ThC (an α-emitter). In particular, it is possible to use an α-β coincidence method (Fontan, 1964; Assaf and Gat, 1967). Fontan's method has been adapted to aircraft measurements.

1) SAMPLING SYSTEM

A yellow quality Schneider-Poelman filter, 20 cm in diameter, collects the atmospheric aerosols. An air flow of 90 m^3 hr^{-1} is obtained by using a 550 W turbine. The collection efficiency is 90%. Depending upon the altitude, the sampling time for each level varies between 5 and 25 min.

2) COUNTING SYSTEM

Filter activity is measured at the laboratory using either an α-scintillation probe or an α-β coincidence system which features a reduced background and counting efficiency (Table 1). The minimum sensitivity, which depends upon the installation used and the sampling time in the airplane, is in the range of 10^{-3} to 10^{-2} pCi m^{-3}. The α-β coincidence installations are used for measuring highest level ThB concentrations because they are more sensitive. When the count rate was low and the measured activity was of the order of the background activity, a least-square method was used to determine the ThB concentrations.

b. Radon measurement

Radon was measured through its daughters which were assumed to be in radioactive equilibrium with the gas in the air. Therefore, the same filter was used for radon and ThB. Measurement of the filter's α-activity was carried out in the airplane immediately after sampling. The counting time varied from 10 to 20 min. A second installation which enables direct measurement of radon by adsorption on active charcoal can also be used. This installation, already described (Guedalia *et al.*, 1972), was not used systematically but only in certain cases.

c. Measurement of meteorological parameters

The airplane used during the experiment campaigns, a TU 2068 Cessna, was equipped by the National Weather Bureau to measure temperature (platinum wire), dew point, relative humidity (carbon plate), static and dynamic pressure, and mean wind direction and speed (Doppler radar). All these parameters were transferred to a photographic recorder. Each vertical profile normally included seven samples. The total time required to perform a vertical profile was always less than 2 hr.

4. Results

a. Comparisons of Rn and ThB measurements

From the total number of vertical profiles, analyses were made only in those cases where the winds were from the west to northwest, that is, the air mass came directly from the ocean. In contrast to other profiles, these cases were characterized by an important decrease in Rn concentration above the mixing layer, showing that Rn advection above the ocean was low. The wind profile and the distance from the coast were taken into

TABLE 2. Vertical exchange coefficients ($m^2 sec^{-1}$) calculated with the infinite and discontinuous source hypotheses in the case of ThB and Rn for air masses coming directly from the Altantic Ocean.

Date	Layer (m)	Mean wind direction (deg)	Mean wind speed ($m sec^{-1}$)	Mean distance to the coast in the direction of the wind (km)	K_z (infinite source hypothesis) [Eq. (4)] ThB	K_z (infinite source hypothesis) [Eq. (4)] Rn	K_z (discontinuous source hypothesis) [Eq. (8)] ThB	K_z (discontinuous source hypothesis) [Eq. (8)] Rn
27/05/72	400–1500	290	15	220	>54	>30	>227	>990
	1500–2000	290	15	220	< 0.3	< 0.4	< 1.6	< 14
29/05/72	300–1000	330	4	380	>21	> 4	> 23	> 20
	>1000	290	5 to 10	220	< 0.16	< 0.015	< 0.26	< 0.1
17/06/72	500–1750	320	4	320	>94	>78	>109	>519
	>1750	290	4 to 7	220	< 0.4	< 0.14	< 0.77	2.3
23/06/72 (05.00)	300– 800	300	8 to 12	230	< 0.4	0.2	< 1	3.3
23/06/72 (17.00)	400–1500	310	7 to 10	250	>17	>12	< 39	>188
	>1500	310	12	250	< 0.1	—	< 0.28	—
11/09/72	300–1650	320	5	320	>88	—	>121	—
	>1650	320	6 to 8	320	< 0.15	0.05	< 0.25	0.6
13/09/72	300–1550	320	7	320	>78	—	>148	—
	>1550	310	9 to 12	250	0.04	0.018	0.08	0.2

account when K_z was calculated using Eq. (8), i.e., zero oceanic concentrations were assumed.

Next, an infinite plane source was assumed and K_z was again calculated using Eq. (4); this exposed the error introduced by that hypothesis. The results are summarized in Table 2.

Figs. 7 and 8 show the profiles obtained for flights made on 17 June 1972 and 29 May 1972, respectively. As in the case of ThB, the rapid decrease in radon concentration above the mixing layer allows the vertical flux to be determined. Moreover, Fig. 8 shows that the mean radon concentration above 1200 m is practically constant and probably corresponds to the concentration existing above the ocean ($\sim$0.4 pCi m^{-3}).

From these experiments, ThB and radon vertical fluxes at 150 m above the ground have been determined. If the amount of tracer contained between the ground and 150 m is ignored, the flux value corresponds to the soil's mean emanation rate between the Atlantic coast and the region where the measurements were made (Table 3). It must be noted that values obtained on 29 May are too low for both Rn and ThB. This is probably the fault of overestimating the continental distance travelled by the airmass. If this value is not considered, the mean value of the radon emanation rate obtained is 8.7×10^{-13} Ci m^{-2} sec^{-1}. ThB concentrations correspond to a mean thoron emanation rate of 1.4×10^{-11} Ci m^{-2} sec^{-1}. The amount of ThB which could deposit on the ground is not considered.

TABLE 3. ThB and Rn vertical flux values at 150 m for air masses coming directly from the ocean. Values in the third and fifth columns result if the infinite source hypothesis is used [Eq. (4)].

Date	Vertical flux at 150 m (pCi $m^{-2}sec^{-1}$) ThB		Rn	
27/05/72	0.02	0.0047	0.66	0.02
29/05/72	0.01	0.009	0.19	0.034
17/06/72	0.019	0.016	0.62	0.094
23/06/72	0.025	0.011	0.68	0.046
11/09/72	0.022	0.016	1.09	0.13
13/09/72	0.039	0.023	1.3	0.12

To facilitate comparison, values for vertical fluxes obtained from the frequently used infinite source hypothesis ($\partial\bar{C}/\partial x=0$) have been indicated in the same table.

In any situation other than that of an air mass moving directly from the ocean, a single radon vertical profile cannot be used to measure the exchange coefficients: 1) the slow decrease in radon concentration in the upper layers makes it impossible to calculate the vertical flux, and 2) without knowing the precise distance covered by an air mass, it is not possible to take the important influence of horizontal advection into account. This latter influence has been shown particularly for air masses reaching Europe from the American continent (Birot *et al.*, 1970; Guedalia *et al.*, 1972).

Fig. 9 shows, as an example, the measurements obtained on 18 March 1972. The temperature profile has

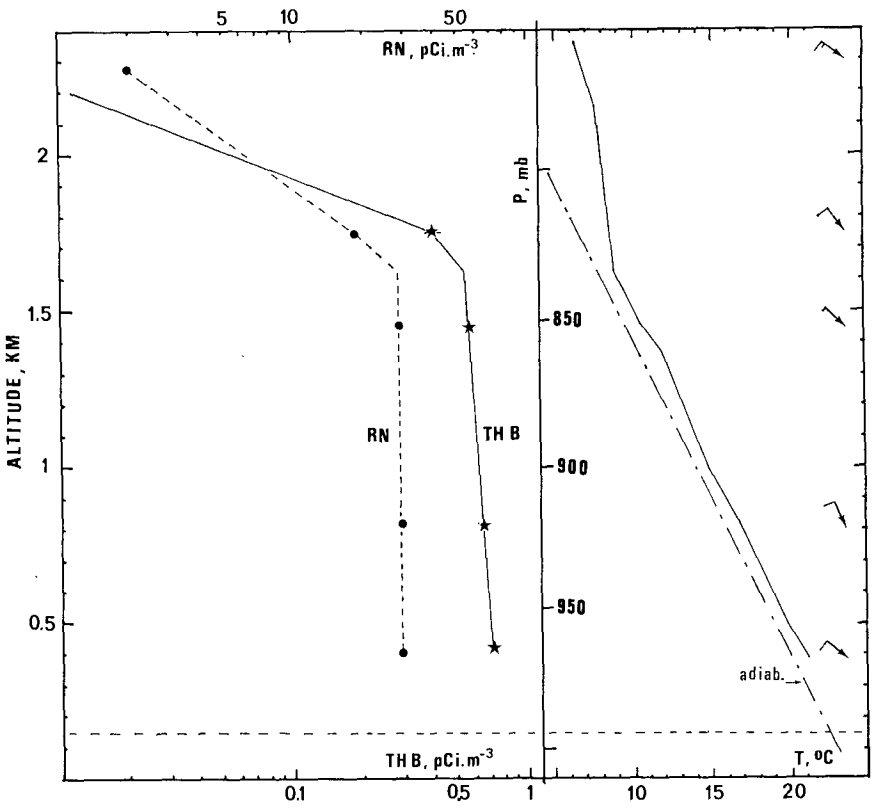

FIG. 7. Vertical profiles of Rn, ThB and temperature corresponding to the flight on 17 June 1972 (ThB concentration at 2300 m <0.06 pCi m^{-3}).

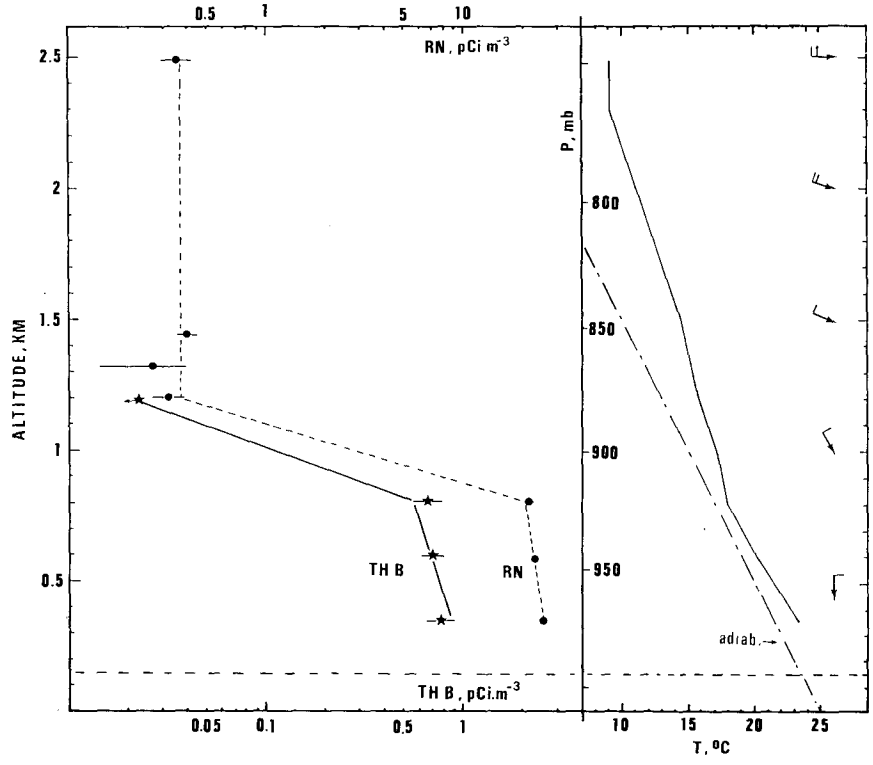

FIG. 8. Vertical profiles of Rn, ThB and temperature corresponding to the flight on 29 May 1972 (ThB concentration at 1200 m <0.02 pCi m^{-3}).

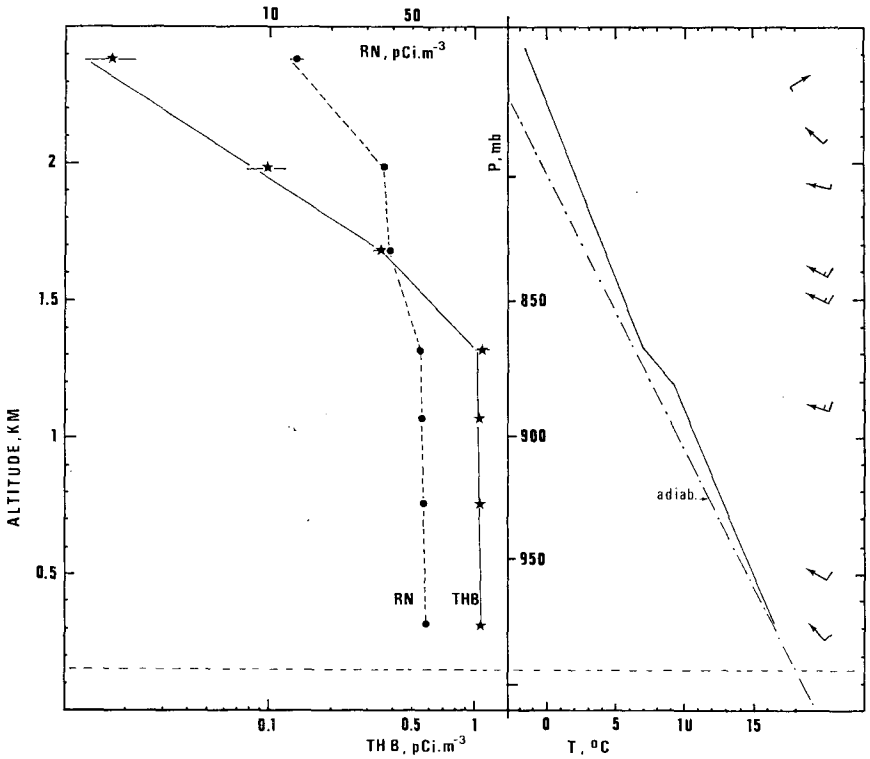

FIG. 9. Vertical profiles of Rn, ThB, and temperature corresponding to the flight on 18 March 1972.

only one lapse rate change at around 1300 m [$\Delta T/\Delta z$ ranges from -1.05 to -0.76C (100 m)$^{-1}$]. A ThB concentration decrease occurs at this level, whereas the Rn concentrations vary only slightly. At the same time, the Rn decrease at 2000 m does not correspond to a diffusivity change but rather to a change in the wind direction; the airflow becomes west-southwesterly, which means that the distance over the continent covered by the air mass is smaller.

To illustrate the steady-state character of the ThB profiles, vertical fluxes for experiments other than those indicated in Table 3 have been calculated. The mean value corresponding to 12 experiments is $(1.7\pm0.8)\times10^{-14}$ Ci m^{-2} sec^{-1} [which corresponds to a Tn emanation rate of $(1.1\pm0.5)\times10^{-11}$ Ci m^{-2} sec^{-1}]. Although it is not possible to calculate the vertical flux values for radon, the total number of Rn atoms contained in an air column is always far less than the number produced by an infinite source equal to the mean ground emanation (Table 3). These results show that the air masses do not remain long over the European continent.

b. Influence of nocturnal stability in the lower layers

ThB and Rn vertical profiles measured on 20 June 1972 at 1630 and those measured the next day at 0530 are shown in Fig. 10. On 20 June the strongly mixed adiabatic layer existing up to 1200 m (all heights MSL) was topped by a temperature inversion between 1200 and 1500 m. The temperature profile above 1200 m for 21 June at 0530 was not very different from that of the previous day but an inversion layer between the ground (150 m) and about 400 m was present. ThB concentrations on 21 June were less by a factor of about 2 than those of the previous day; the strong stability in the lower layers considerably reduced the vertical flux at 400 m and the decrease in concentration probably corresponded to radioactive decay. Rn concentrations changed little from one day to the other. As a matter of fact, Rn's long half-life prevents it from being very sensitive to daily diffusivity variations, above several hundred meters. It must also be pointed out that the radon distribution does not depend upon local diffusivity conditions. An increase in radon with altitude brought about by horizontal advection can thus be noted.

Whenever possible, several temperature soundings were made before the samples were taken, thus making it possible to determine lapse rate changes, particularly during the period when the nocturnal stability in the lower layers disappears, a situation which precipitates sudden changes in vertical diffusivity. It is, therefore, more or less certain that ThB profiles are in steady-state conditions.

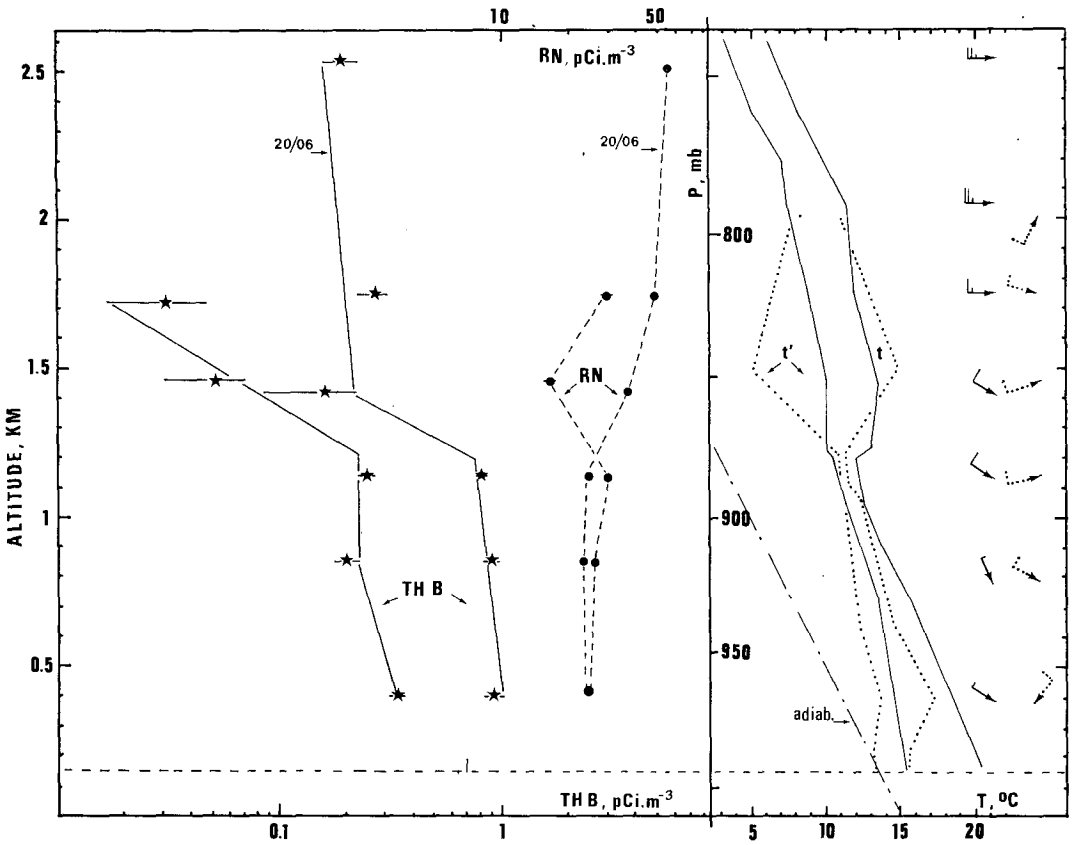

FIG. 10. Vertical profiles of Rn, ThB and temperature corresponding to the flight on 20 June 1972 (1700) and on 21 June 1972 (0500). Temperature (t), wet bulb (t') and wind profiles for 21 June are given by dotted lines.

c. Relationship between the temperature gradient and the exchange coefficients

The exchange coefficients calculated from all the ThB profiles are shown in Fig. 11 as functions of the temperature gradient Three cases can be considered:

1) VERY UNSTABLE LAYERS $[\Delta T/\Delta z < -0.95\text{C (100 m)}^{-1}]$

During daytime these layers very often reach from the ground up to several hundred meters. They are the site of strong vertical mixing caused by thermal instability. Since it has been shown in this case that the ThB gradient cannot be measured, only a lower limit of the exchange coefficient has been calculated. The smallest computed lower limits ($K_z=21$ and $K_z=39$) correspond to two experiments in which the experimental error in the measurement is important and leads to an increase in the gradient used to calculate K_z. It can be established that in these layers, the exchange coefficients are greater than 100 m^2 sec^{-1}. The dispersion of K_z values does not seem to be related to various exchange conditions but rather to experimental factors (thickness of the layer considered, experimental errors, etc.). It is interesting to note that it is radon which unexpectedly allows the highest K_z values to be determined in the case of an oceanic air mass traveling a short distance over the continent. Thus, it can be seen by referring to Table 2 that the highest K_z value calculated using radon is greater than 990 m^2 sec^{-1}. Values obtained with radon are not shown in Fig. 11 since in that case an error in determining $\bar{u}$ and x would induce an important error in determining K_z.

2) STABLE LAYERS $[\Delta T/\Delta z \geqslant -0.65\text{C (100 m)}^{-1}]$

In this case, with larger temperature gradients, the development of vertical motion is significantly retarded. Generally, the presence of a stable layer above the planetary boundary layer can be observed during daytime. During nightime, a stable layer often develops near the ground (nocturnal stability). Fig. 11 shows that in spite of some dispersion, the lowest K_z values correspond to the strongest temperature inversions. The lowest K_z value (8×10^{-2} m^2 sec^{-1}) is obtained for a temperature gradient of 3.6C (100 m)$^{-1}$. But whatever the $\Delta T/\Delta z$ value is, K_z is always less than 5 m^2 sec^{-1}. Possible wind influence on the vertical exchange could

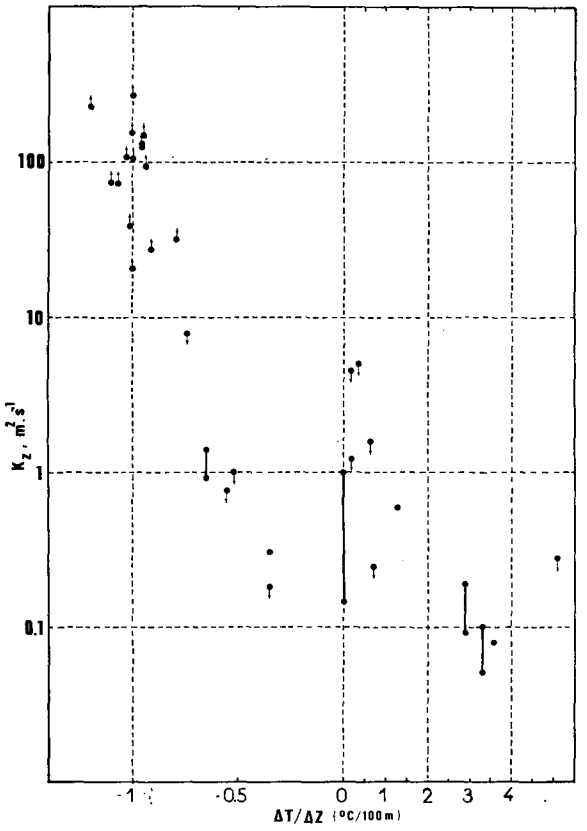

FIG. 11. Variations of K_z, calculated using ThB, as a function of the temperature gradient.

not be studied with this group of results, for there were not enough data collected to obtain significant statistics.

3) INTERMEDIATE LAYERS $[-0.95<\Delta T/\Delta z<-0.65\text{C}\ (100\ \text{m})^{-1}]$

First, it can be noted that there are only a small number of cases corresponding to these temperature gradients. For a gradient less than $-0.75\text{C}\ (100\ \text{m})^{-1}$, K_z is greater than 30 m² sec⁻¹, while for the only gradient between -0.75 and $-0.65\text{C}\ (100\ \text{m})^{-1}$, K_z is less than 8 m² sec⁻¹. It seems, therefore, that in this transition zone the exchange coefficients vary rapidly with the temperature gradient. These layers can be considered as a transition between the stable layers and the intense vertical mixing layers.

5. Conclusion

The various methods developed in this paper make it possible to use Rn and ThB vertical profiles to calculate vertical exchange coefficients in the planetary boundary layer and between this layer and the free atmosphere. The geographic position of France is such that there are frequent encounters of oceanic air masses. When this situation occurs, the ocean-continent discontinuity of the source is taken into account in calculating vertical exchange coefficients.

The variation (between 10^{-1} and several 100 m² sec⁻¹) of the vertical exchange coefficients is important and is closely related to the temperature gradient. Coefficients determined in this way are characteristic of the aerological scale. It was not possible, however, to determine the influence of the wind (for a given temperature gradient) on the exchange coefficients.

It is interesting to note that the development of a turbulent layer starting from the ground during daytime leads to the formation of a stable zone and, fairly often, to a temperature inversion between the mixing layer and the free atmosphere. In this situation the inversion could disappear when turbulence disappears, that is, during nightime; and at that time, the exchange between the lower layer and the free atmosphere can increase. However, it often happens that the inversion caused by diurnal turbulence blends with a synoptic inversion (e.g., a subsidence inversion). In this case the disappearance of this stable zone between the planetary boundary layer and the free atmosphere is not related to the daily cycle.

The low vertical exchange coefficients corresponding to temperature inversions ($K_z \approx 0.1$ m² sec⁻¹) show that the vertical exchanges resulting from turbulent diffusion between the planetary boundary layer and the free atmosphere are often relatively small. A good knowledge of the relationship between the temperature gradient and K_z should allow us to evaluate the vertical exchanges when the vertical temperature gradient is known.

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