

Lead-212, radon and vertical mixing in the lower Atmosphere (100–2 000)m

By D. GUEDALIA, C. ALLET, J. FONTAN and A. DRUILHET, *Université Paul Sabatier, Toulouse, France* and G. ASSAF, *Weizmann Institute of Science, Rehovot, Israel*

(Manuscript received June 9, 1972; revised version February 8, 1973)

ABSTRACT

This paper summarizes 5 vertical soundings of lead-212 concentrations in the lower atmosphere, in 3 of which lead was measured together with radon. The measurements were made in stable weather and under light winds conditions in the South West of France. There was general agreement among radon and lead and the stability of the atmosphere, which indicate a semi-steady state situation. Using radon and lead profiles we use an advection diffusion model to estimate the mean vertical motion and the vertical diffusion. The results indicate downward wind of less than 1 cm/sec and vertical mixing which is very sensitive to stability. For Brunt-Väisälä frequency $N^2 = 3.5 \times 10^{-4} \text{ sec}^{-2}$ which is characteristic of isothermal atmosphere $K \approx 1 \text{ m}^2/\text{sec}$. For a profile which is half adiabatic ($N^2 = 1.7 \times 10^{-4} \text{ sec}^{-2}$) $K = 10 \text{ m}^2/\text{sec}$, where N^2 is smaller than $5 \times 10^{-5} \text{ sec}^{-2}$, K is greater than $40 \text{ m}^2/\text{sec}$. These estimates were obtained on different days for levels of similar stabilities.

Introduction

There are very few observations on the nature of vertical mixing in the atmosphere above 100 m or so. Some attempts were made to estimate eddy viscosity from the wind profiles (Lettau, 1952; Blackadar, 1962). The dynamic methods can be applied when geostrophic flow is modified by the boundary through the effect of eddy viscosity, i.e., in the friction layer. In stable stratification and above the friction layer it is generally very difficult, if not impossible, to relate the mean wind profile to eddy viscosity.

Smoke plumes were successfully used by Byzova et al. (1970) for studying atmospheric microstructure. This application, however, is limited to small-scale phenomena only.

So far most of the attempts to apply emanation products to the study of atmospheric transport were based on measurements of radon (Kirichenko, 1964; Bradley & Pearson, 1970; Jacobi & Andre, 1963). Yet theory, experiments and numerical studies indicate that radon response primarily to the large scale of atmospheric motion (Assaf, 1969, 1971; Birot et al., 1970; Guedalia et al., 1971; Huss et al., 1970; Nazarov et al., 1970). This occurs to a

smaller degree with Pb^{212} . Its 10.6 h half life is long enough to integrate the microscale effects and its distribution is less sensitive to the macroscale effects of atmospheric motion (Assaf, 1969). In this paper we describe an attempt to utilize Pb^{212} for the study of atmospheric mixing throughout the boundary layer in the south-west of France.

Experimental procedure and weather conditions

The samples were taken aboard an aircraft together with temperature recordings. Air was pumped through filter paper at a rate of $80 \text{ m}^3/\text{h}$ for 10 to 20 min. In October 1969 the filters were introduced into liquid scintillation chamber and analysed by a delayed coincidence counter (Assaf & Gat, 1966). In 1971 we utilized the coincidence technique of Fontan et al. (1962).

The radon was measured by α -counting aboard the plane. This method was compared with measurements of radon trapped by absorption of an activated charcoal (Delos, 1971). The flights were made about 30 km to the west of Toulouse.

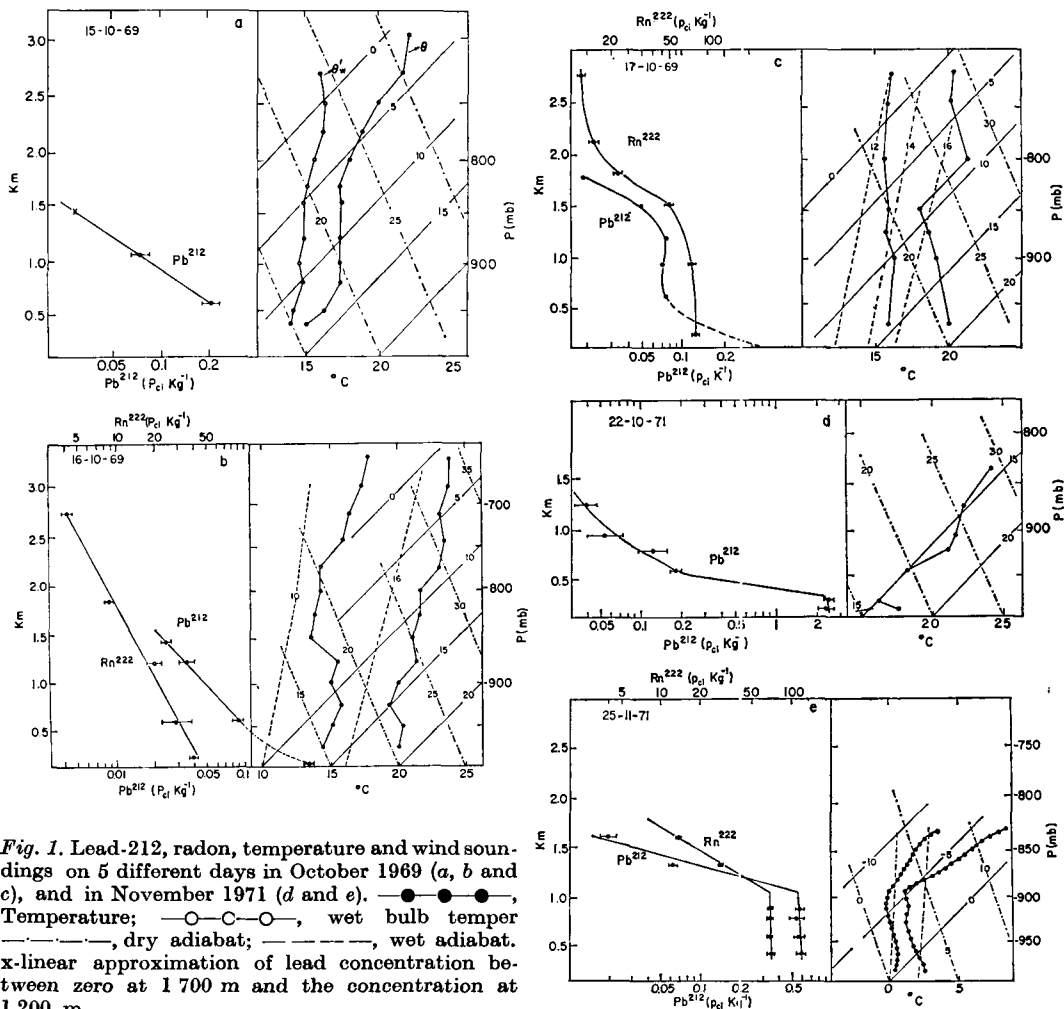


Fig. 1. Lead-212, radon, temperature and wind soundings on 5 different days in October 1969 (a, b and c), and in November 1971 (d and e). —●—●—●—, Temperature; —○—○—○—, wet bulb temp; — — —, dry adiabat; — — —, wet adiabat. x-linear approximation of lead concentration between zero at 1 700 m and the concentration at 1 200 m.

Fig. 1 presents the concentration profiles of Pb^{212} (or lead) together with radon, temperature and wind sounding from Toulouse airport. On 15.10.69 high pressure developed over Northern France and low pressure over Corsica. The winds were light and in varying directions. On 16 October the high pressure developed all over Western Europe and remained so on the 17th of October. Also on October 22nd 1971 high pressure and light winds characterized the weather over the South of France. On November 11, 1971 the low pressure was centered over Western Mediterranean and high pressure over the Bay of

Biscay. Thus, altogether the soundings were taken in stable conditions, winds at the surface and the 850 mb generally did not exceed 10 knot, and the stronger wind measured in the flight on October 17th and 16th were of local origin in Toulouse only as we did not find such wind on the synoptic charts.

We noted that on the 16th of October the sampling area was closer to the centre of high pressure (surface pressure was 1022 mb), while on the 17th we sampled over the periphery of the high pressure (surface pressure dropped to 1 015 mb).

Result and discussion

Looking at Fig. 1 one can notice that there is general agreement between the stability and the distribution of Pb²¹² and radon. Thus whenever the stability is close to neutral the distribution of the tracers is very uniform with height. We use a Brunt-Väisälä frequency

$$N^2 = \frac{g}{\theta} \frac{d\theta}{dz}$$

as an index of stability and related the characteristic heights of Pb²¹² and radon and the apparent diffusivities to this index (Table 1) (the apparent diffusivities were defined as $K_a = (H^2\lambda)$ where H is the characteristic height in which the concentration decreases by a factor of e and λ the decay constant).

In the two consecutive days, October 16th and 17th, when radon and lead were measured, the integrated amount of lead remained the same—13 DPS/m²—while that of radon changed from 260 to 460 DPS/m². However, the correspondence between the radon, lead and temperature profiles (Fig. 1) and the fact that the apparent diffusivities of radon are consistently about half those of lead (Table 1), suggest that on these particular days even if radon was imported to the area by horizontal advection, it had stayed long enough in atmosphere of similar characteristics.

As our measurements were carried out in high pressure regimes, we expect that the apparent diffusivities from radon and lead will both be an underestimate of the vertical

diffusion. In Table 1 one can see that in general there is agreement between the stability and the apparent diffusivities. Also, the apparent diffusivities from radon are somewhat smaller than those derived from Pb²¹² as expected in downward wind (Assaf 1969). This suggests an application of advection diffusion model for our measurements.

Nir (1964) has shown that in advection diffusion systems the characteristic distance H_i in steady state and for one dimension can be expressed as:

$$H_i = \frac{2K}{(W^2 + 4\lambda_i K)^{1/2} - W} \quad (1)$$

where i is the index of radon or lead profile, W is the mean velocity and λ_i the decay constant of the specific radio isotope (see Assaf, 1969, for extension of (1) to 3 dimensions).

We solve these equations for W and K using the profiles that radon was measured together with lead. The results are given in Table 1.

The last two columns of Table 1 give the nondimensional numbers $A_i = W/(\lambda_i K)^{1/2}$ which are the indicator of the relative importance of diffusion versus advection. When A_i is bigger than 1, advection is the dominant mode of transport, and when it is smaller than 1, diffusion becomes dominant (Assaf, 1969). One can note that $A(\text{Rn})$ is bigger and $A(\text{Pb})$ smaller than 1; this shows that lead responded primarily to diffusion, while radon to advection in all three relevant soundings.

Thus the estimates of K are essentially based upon the lead, and that of W upon radon distribution.

Table 1.

Date	Layer (m)	N^2 (sec ⁻²)	H_{Pb} (m)	$K_a(\text{Pb})$ (m ² /sec)	H_{Rn} (m)	$K_a(\text{Rn})$ (m ² /sec)	W (cm/sec)	K (m ² /sec)	$A(\text{Pb})$	$A(\text{Rn})$
15.10.69	600–1 200	$2.10 \cdot 10^{-4}$	500	5.	—	—	—	—	—	—
16.10.69	600–1 400	1.7×10^{-4}	650	7	1 300	3.5	–0.7	11	0.5	1.5
	100–1 800	1.7×10^{-4}	—	—	1 300	3.5	—	—	—	—
17.10.69	600–1 500	5.2×10^{-5}	∞	∞	∞	∞	—	—	—	—
	1 500–1 800	3.5×10^{-4}	250	1.3	430	.4	–0.43	2.5	0.65	2.0
	1 800–2 800	5×10^{-5}	—	—	4 200	40	—	—	—	—
22.10.71	200– 300	$N^2 = 0$	∞	∞	—	—	—	—	—	—
	300– 550	3.5×10^{-4}	109	0.2	—	—	—	—	—	—
	550–1 200	3.5×10^{-4}	370	2.4	—	—	—	—	—	—
25.11.71	200–1 100	3.3×10^{-5}	∞	∞	—	—	—	—	—	—
	1 100–1 600	5×10^{-4}	155	0.4	325	0.2	–0.14	0.6	0.4	1.3

Looking at Table 1 we notice that stability remained the same in the level 600–1 200 m on the 15th and 16th of October 1969. The apparent diffusivities $K_a(\text{Pb})$ was also similar, 5. and 7. m^2/sec . On the 16th the corrected K value gives 11 m^2/sec and on the 15th we did not measure radon, so we could not correct for the vertical motion. At any rate, we have good reason to believe that on the 16th lead distribution was close to steady state and the value of 10 m^2/sec represents the vertical mixing for the half adiabatic rate where $N^2 = 1.7 \times 10^{-4} \text{ sec}^{-2}$.

The minimum value of vertical diffusion $H^2\lambda = 0.2 \text{ m}^2/\text{sec}$, was found on October 22nd, 1971, between 300 to 550 m. We think that this low value is due to the erosion from below of the stable layer, as indicated from the temperature soundings (Fig. 1). It appears that for the same stability ($N^2 = 3.5 \times 10^{-4} \text{ sec}^{-2}$) $H^2\lambda = 2.4 \text{ m}^2/\text{sec}$ on the same day, but at a different altitude (550–1 200 m). Also, on October 17th 1969, we obtained $K = 2.5 \text{ m}^2/\text{sec}$ for the similar stability, and we think this value is representative of isotherm atmosphere with light winds. For $N^2 < 5 \times 10^{-5}$ the vertical diffusion approximates 40 m^2/sec or more. Actually the slope of lead and radon distribution in those conditions is so small that K appears to be infinite. It is interesting to note that in conditions close to neutral over urban boundary layer, Assaf & Biscaye (1972) reported eddy diffusion of the same magnitude.

The validity of the results

In general a quantitative estimate of the vertical motion in the lower atmosphere should be based upon a net of radon sounding extending to an area comparable to the size of the synoptic system (Huss et al., 1970). Our estimates of W should be looked on only as an indicator of magnitude. Much more can be said about the estimates of K .

The remarkable correspondence between lead and temperature profiles and the integrated amount of lead that remains the same for the two consecutive days in October 1969 (on the 15th of October we did not take any measurements near the surface and we could not estimate the integrated amount of lead) indicate that lead concentration reflects steady state

conditions in which the decay is balanced by local vertical transport. As lead concentration is not very sensitive to vertical advection, the estimate of K which is based essentially upon the distribution of lead seems to be meaningful.

Eq. 1 assumes also the validity of the K approximation. This assumption should be considered with caution. It was shown by Assaf (1969) that instantaneous measurements of radio isotopes ("phase mean" in the reference) integrate the effects of eddies which are short-lived, compared to the isotopic mean life into the apparent eddy diffusion K_a (see also Beran & Assaf, 1970). In our case, when there is intensive activity of eddies with mean lives comparable to the 15 hour mean life of lead, $K_a(R_n)$ will be very different from $K_a(P_b)$. Radon will "see" those eddies as diffusion elements and their effect will be incorporated into $K_a(R_n)$. However, these eddies will make lead concentration nonsteady and the $K_a(P_b)$ will change its magnitude according to the phase of these eddies at the time of measurement. The general agreement between $K_a(\text{Pb})$ and $K_a(\text{Rn})$ indicates that at the time of the measurements there was not much eddy activity in that range.

Another process that should be taken into account in evaluating Table 1 is the erosion of the stable layer. This process that occurred between the 16th and 17th of October in the lower atmosphere resembles the erosion of the thermocline by mechanical action of the wind. The mean lapse rate of temperature between 200 and 2 000 m remained the same, yet on the 17th, the first 1 400 m became well mixed according to lead, radon and temperature sounding. We can deduce that the gradient above the mixed layer will increase and the apparent diffusivities derive from lead and radon will underestimate the existing conditions similar to the case of the erosion of the thermocline in the ocean. We expect that the lead and radon gradient will be small and the apparent diffusivities will be an overestimate of the instantaneous mixing if we sample just after the onset of stability in the lower atmosphere. We also expect that the lead concentration will adjust itself sooner than the radon because of the decay.

Acknowledgements

We would like to thank Y. Lapid and A. Birot for participating in the measurements of radon and lead in our flights.

REFERENCES

- Assaf, G. 1969. Emanated products as a probe of atmospheric transport. *Tellus* 21, 820.
- Assaf, G. 1971. Comments of a paper by W. E. Bradley & J. E. Pearson, "Aircraft measurements of the vertical distribution of radon in the lower atmosphere". *J. Geophys. Res.* 76, 2897.
- Assaf, G. & Biscaye, P. E. 1972. Pb²¹² in the New York City urban boundary layer. *Science* 175, 890.
- Birot, A., Adroguer, B. & Fontan, J. 1970. Vertical distribution of Rn-222 in the atmosphere and its use for study of exchange in the lower troposphere. *J. Geophys. Res.* 75, No. 12, 2373.
- Beran, M. & Assaf, G. 1970. Use of isotopic decay rates in turbulent dispersion studies. *J. Geophys. Res.* 75, 5279-5285.
- Blackader, A. K. 1962. The vertical distribution of wind and turbulent exchange in neutral atmosphere. *J. Geophys. Res.* 67, 3095.
- Bradley, W. E. & Pearson, J. E. 1970. Aircraft measurements of the vertical distribution of radon in the lower atmosphere. *J. Geophys. Res.* 75, 5890.
- Byzova, N. L., Ivanov, V. N., Garger, E. K. & Osipov, Y. S. 1970. Use of statistical wind characteristics for theoretical and experimental estimation for passive particle diffusion. *J. Geophys. Res.* 75, 3605.
- Delos, Y. 1970. Detection du radon dans la mer et dans l'atmosphère. These No. 932, Toulouse.
- Fontan, J., Blanc, D & Bouville, A. 1962. Emploi d'un système de coïncidences dans le dosage sélectif du Rn et du Tn atmosphérique. *J. de Phys. et Rad.* 23, 333.
- Gat, J. R. & Assaf, G. 1968. Atmospheric Bi²¹². Measurements and some geophysical applications. *Science* 159, (3818) 977.
- Guedalia, D., Birot, A., Fontan, J. & Allet, C. 1971. Sur la détermination des coefficients de diffusion dans la couche limite planétaire à l'aide du radon et du ThB. *C. R. Acad. Sci. Paris*, t. 273, p. 57.
- Huss, A., Segal, M. & Assaf, G. 1970. Model investigation of time dependent distribution of radon. *J. Geophys. Res.* 75, 3020.
- Jacobi, W. & Andre, K. 1963. The vertical distribution of radon-222 and radon-220 and their decay products in the atmosphere. *J. Geophys. Res.* 68, 3799.
- Kirichenko, L. V. 1964. Vertical distribution of the products of decay of radon in the free atmosphere, problems in nuclear meteorology. AEC tr. 6128, 92.
- Lettau, J. 1950. Re-examination of the "Leipzig wind profile", considering some relations between wind and turbulence in the frictional layer. *Tellus* 2, 125.
- Nazarov, L. E., Kuzenkov, A. F., Malakhov, S. G., Volokitina, L. A., Gaziyeve, Ya. I. & Vasil'yev, A. S. 1970. Radioactive Aerosol Distribution in the Middle and Upper Troposphere over the USSR in 1963-1968. *J. Geophys. Res.* 75, 3575.
- Nir, A. 1964. On the interpretation of tritium "age" measurements of groundwater. *J. Geophys. Res.* 69, 2589.

СВИНЕЦ-212, РАДОН И ВЕРТИКАЛЬНОЕ ПЕРЕМЕШИВАНИЕ В НИЖНЕЙ АТМОСФЕРЕ (100-2 000 м)

В статье суммируются результаты 5 вертикальных зондирований концентраций свинца-212 в нижней атмосфере, в трех из которых свинец измерялся вместе с радоном. Измерения производились при устойчивой погоде в условиях легкого ветра на юго-западе Франции. Наблюдалось общее соответствие между концентрациями свинца и радона и устойчивостью атмосферы, что указывает на квазистационарность ситуации. Используя профили концентраций свинца и радона, мы с помощью адвективно-диффузионной модели оцениваем средние вертикальные движения и

диффузию. Результаты указывают на нисходящие вертикальные движения со скоростями, меньшими 1 см/сек, и на то, что вертикальное перемешивание очень чувствительно к устойчивости. Для частоты Брента-Вяйсяля $N^2 = 3,5 \cdot 10^{-4} \text{ сек}^{-2}$, что характерно для изотермической атмосферы, $K \approx 1 \text{ м}^2/\text{сек}$. Для профиля, градиент температуры в котором равен половине адиабатического ($N^2 = 1,7 \cdot 10^{-4} \text{ сек}^{-2}$), $K \approx 10 \text{ м}^2/\text{сек}$, в случаях, когда N^2 меньше $5 \cdot 10^{-5} \text{ сек}^{-2}$, K больше $40 \text{ м}^2/\text{сек}$. Эти оценки получены в разные дни для аналогичных условий устойчивости.