The Vertical Distribution of Radon 222, Radon 220 and Their Decay Products in the Atmosphere

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Abstract. The vertical equilibrium distributions of Rn^{222}, Rn^{220}, and their decay products in a horizontally isotropic atmosphere are calculated from diffusion theory with the help of a digital computer. The change of the turbulent diffusion coefficient with altitude and the removal of Pb^{210} and Po^{210} by fallout were taken into account. Vertical profiles of the natural radionuclides in the atmosphere are presented for several typical profiles of the turbulent diffusion coefficient. Agreement with available experimental data is rather good. The results indicate the importance of the natural radioactive tracers in the study of mixing processes in the troposphere and in the mechanism of transport from the troposphere into the lower stratosphere.

Introduction

Our knowledge of the processes of diffusion of aerosols in the atmosphere and removal from it is rather limited, although these processes are of considerable importance in estimating the hazards following the release of radioactive and inactive toxic materials in the free air. Rn^{222}, Rn^{220}, and their decay products are valuable, natural tracers for the study of these processes in the troposphere and lower stratosphere, but except for the boundary layer near ground level only a few measurements of Rn^{222}, Pb^{210}, and Po^{210} are available at present.

For a general view of the vertical distribution of Rn^{222}, Rn^{220}, and their decay products in the atmosphere under different conditions of turbulence, theoretical computations on the basis of the exchange theory are useful. Previous attempts in this direction were made by *Hess and Schmidt* [1918], *Schmidt* [1926], *Priebsch* [1932], *Philip* [1959], and *Malakhov* [1959], who calculated the vertical distribution of Rn^{222} assuming a constant value or a power law for the increase of the vertical diffusion coefficient with altitude. Under these assumptions analytical solutions of the diffusion equation for Rn^{222} and Rn^{220} are possible, but they represent only rough approximations to the real conditions in the atmosphere.

For more realistic results we solved the diffusion equation by a numerical method which enabled us to use any vertical profile of the turbulent diffusion coefficient. Furthermore, this method can be applied to all decay products of Rn^{222} and Rn^{220} by taking into consideration their removal from the atmosphere by diffusion and washout. The computations involved in this method are considerable. They could be done only on a fast electronic computer.

The Diffusion Equation and Its Boundary Conditions

The spreading out of Rn^{222} and Rn^{220} in the atmosphere after their exhalation from the ground is caused by turbulent diffusion and convection and is limited only by the radioactive decay of the two nuclides. In contrast to the emanations, the decay products, being isotopes of heavy metals, become readily attached to aerosol particles and precipitation elements. The distribution of these decay products is thus controlled not only by radioactive decay and by diffusion but also by sedimentation and washout, i.e., by the removal of aerosols to the earth's surface.

Because of these processes the concentration of a nuclide in the decay chain at a point having horizontal coordinates x and y at an altitude z above ground level ($z = 0$) is given by the general expression

$$\frac{\partial n_i}{\partial t} = \nabla \cdot (K \nabla n_i) - \mathbf{u} \cdot \nabla n_i$$

$$+ v_i \frac{\partial n_i}{\partial z} + \lambda_{i-1} n_{i-1} - (\lambda_i + \Lambda_i) n_i \qquad (1)$$

where

$i =$ the index for the position of the nuclide in the decay chain (Rn^{222}, Rn^{220} : $i = 1$; Po^{218}, Po^{216} : $i = 2$; $\cdots$)

$u_x, u_y, u_z =$ the components of the mean wind velocity.

$K =$ the turbulent diffusion coefficient.

$v_i =$ the mean sedimentation velocity of the ith nuclide, according to sedimentation of their carrier particles (Rn^{222}, Rn^{220} : $v_1 = 0$).

$\lambda_i =$ the radioactive decay constant of the ith nuclide.

$\Lambda_i =$ the mean removal rate the ith nuclide, caused by washout and rainout (Rn^{222}, Rn^{220} : $\Lambda_1 = 0$).

The terms on the right side of (1) refer, in this order, to turbulent mixing, convection, sedimentation of radioactive particles, radioactive buildup from the mother nuclide in the decay chain, radioactive decay, and removal by washout and rainout.

We are primarily interested in the mean vertical distributions for steady-state conditions at constant exhalation rates of Rn^{222} and Rn^{220}. These equilibrium profiles are obtained from (1) if $\partial n_i / \partial t = 0$. To solve (1) we make the following additional assumptions:

1. $\partial n_i / \partial x = \partial n_i / \partial y = 0$. This means a horizontal isotropic distribution, which will occur when the vertical turbulence profile and the Rn^{222} and Rn^{220} exhalation rates are the same at each place.

2. $u_z = 0$. A reasonable mean value of the vertical wind velocity cannot be given, but in most cases it will be small compared with the velocity of vertical turbulent diffusion.

3. $v_i = 0$. The mean radius of natural radioactive particles in the atmosphere is smaller than $1\ \mu$; the corresponding sedimentation velocity is smaller than about 1 m/h, i.e., small compared with the transport velocity caused by turbulent mixing.

4. $\Lambda_i =$ constant $= \Lambda$. This means that the removal rate is independent of altitude and equal for all decay products. The independence of altitude will be approximately true in the troposphere, whereas above the tropopause Λ will be zero. This assumption is of importance only for the distribution of Pb^{210} and Po^{210}, be-

cause for the short-lived Rn^{222} decay products and all Rn^{220} decay products $\lambda_i \gg \Lambda_i$. For Pb^{210} and Po^{210} nearly the same removal rate can be expected.

With these assumptions the equilibrium vertical profiles of Rn^{222}, Rn^{220}, and their decay products can be obtained from the following system of differential equations:

$$\frac{d}{dz}\left(K \frac{dn_1}{dz}\right) - \lambda_1 n_1 = 0$$

$$\frac{d}{dz}\left(K \frac{dn_i}{dz}\right) + \lambda_{i-1}n_{i-1} - (\lambda_i + \Lambda)n_i = 0$$

$$\text{for } i > 1 \qquad (2)$$

To solve (2), we introduce the following boundary conditions:

1. $\int_0^\infty \lambda_1 n_1 dz = E_1$ where E_1 is the exhalation rate of Rn^{222} and Rn^{220} atoms from the ground surface ($z = 0$). This condition means that the total activity of Rn^{222} and Rn^{220} in a vertical air column is equal to the exhalation rate of its ground-surface area.

2. $E_i = 0$ for $i > 1$. This condition reflects the rapid diffusion of newly formed Po^{218} or Po^{216} atoms in ground air and their deposition on the ground material.

3. $n_i(z = 0) = 0$ for $i > 1$. This takes into account the fact that all atoms and carrier particles of the decay products reaching the ground surface by diffusion will be deposited.

4. $n_i(z \to \infty) \to 0$ for $i = 1, 2, 3, \ldots$. This condition is a consequence of radioactive decay.

The diffusion coefficient K is quite variable with altitude according to the vertical variations of wind velocity and atmospheric stability. Some characteristic air layers can be distinguished from the relative slope of the function $K(z)$. In the boundary layer near the earth's surface K increases rapidly with altitude, following an approximately linear or power law of z. Approaching the gradient wind height, dK/dz decreases and becomes nearly independent of altitude in the upper troposphere under normal conditions. Owing to the high stability of the stratosphere, K again decreases rapidly above the tropopause and is likely to approach a rather constant value in the lower stratosphere.
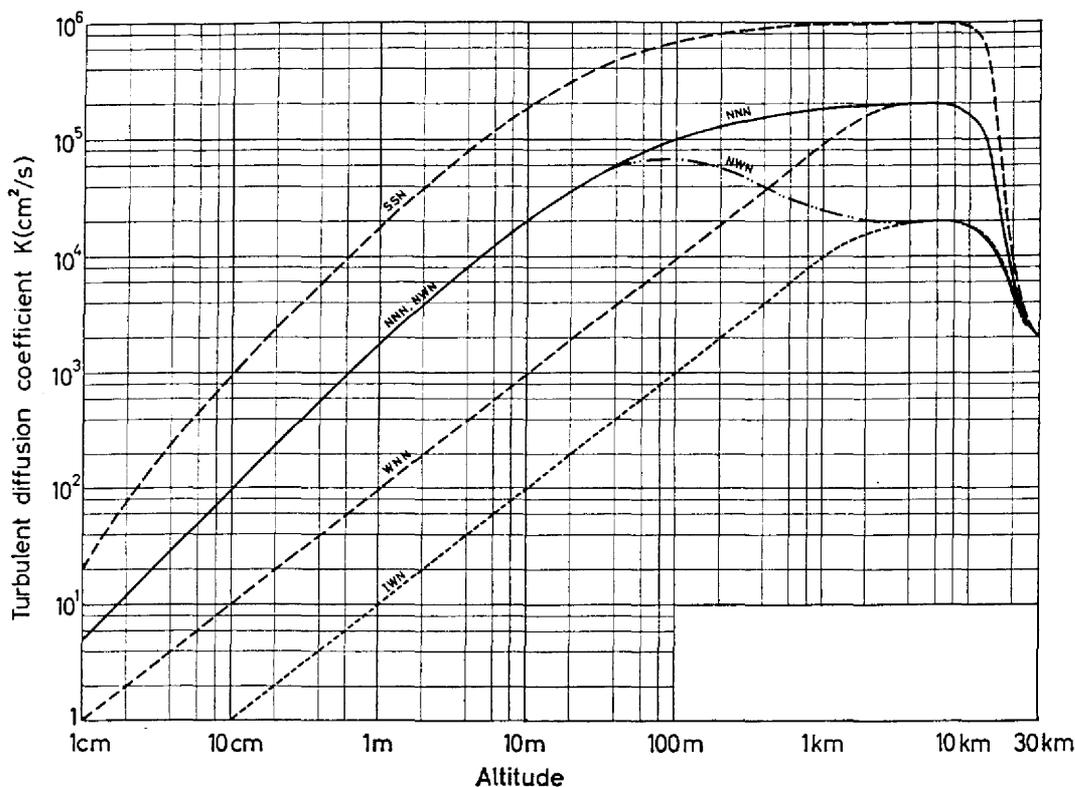
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Fig. 1. Vertical profiles of the turbulent diffusion coefficient.

profiles with time result in fluctuation of K by about 2 or 3 orders of magnitude in the ground layer and by about 1 or 2 orders of magnitude in the upper troposphere. The influence of these uncertainties on the vertical distribution of the natural radionuclides was estimated from solutions of (2) for twenty different K profiles. These profiles cover the total range of fluctuation of the turbulent diffusion coefficient that seems to be possible according to the diffusion diagram given by *Lettau* [1951].

Five characteristic K profiles which correspond to typical conditions of turbulence in the troposphere are shown in Figure 1. The K profile *SSN* is an example of strong vertical mixing, and the profile *WNN* is an example of rather weak vertical mixing throughout the lower troposphere. For *IWN*, diffusion in the ground layer was assumed to be very weak, which is typical for a strong inversion near ground. The profile *NNN* is likely to be representative of normal turbulence conditions throughout the troposphere. In all these five typical cases a

rapid decrease of the turbulent diffusion coefficient was assumed in the tropopause region approaching a constant value of $K = 3 \times 10^4$ cm²/ sec between 20 and 30 km. In either case the gas kinetic diffusion coefficient, which is about 0.05 cm²/sec for Rn^{222} and Rn^{220} atoms, can be neglected.

NUMERICAL TREATMENT

We use the abbreviations

$$\lambda_\nu^* = \lambda_1 \quad \text{for} \quad \nu = 1$$
$$= \lambda_\nu + \Lambda \quad \text{for} \quad \nu > 1$$

Putting

$$\varphi_1 = n_1, \qquad \varphi_i = n_i + \sum_{\nu=1}^{i-1} \alpha_{i,\nu} n_\nu$$

$$i > 1 \quad (3)$$

and defining $\alpha_{i,\nu}$ recursively by

$$\alpha_{i,\nu} = \frac{\lambda_\nu}{\lambda_\nu^* - \lambda_i^*} \alpha_{i,\nu+1}$$

$$\nu = 1, \ldots, i - 1 \quad (4)$$

and

$$\alpha_{i,i} = 1$$

we obtain from (2) the following homogeneous system of differential equations for the φ_i:

$$(K\varphi_i')' = \lambda_i^* \varphi_i \qquad (5)$$

The boundary conditions for $i = 1$ are

$$\varphi_1(\infty) = 0$$

$$K(0)\varphi_1'(0) = -E_1$$

and for $i > 1$ they are

$$v_i(\infty) = 0$$

$$v_i(0) = \alpha_{i,1}v_1(0)$$

After evaluating $\varphi_1, \varphi_2, \cdots$ successively, we determined the functions n_i from (3). For the evaluation the exhalation rates of Rn^{222} and Rn^{220} were standardized to $E = 1$ atom/cm² sec.

For the numerical treatment the given K profile was approximated by a step function. If $z_0 = 0, z_1, \cdots, z_n$ are the points of discontinuity of K, and if $K(z) = K_j$, for $z_j \leq z < z_{j+1}$, the φ_i are linear combinations of $\exp(\beta_j z)$ and $\exp(-\beta_j z)$, where $\beta_j = (\lambda_r/K_j)^{1/2}$, when z is restricted to an interval in which K is constant. φ_i and $K\varphi_i'$ must remain continuous at the points of discontinuity of K.

In particular,

$$\varphi_i'(z_j - 0) = (K_j/K_{j-1})\varphi_i'(z_j + 0) \qquad (6)$$

where $\varphi_i'(z_j - 0)$ and $\varphi_i'(z_j + 0)$ denote left and right limits, respectively.

An easy computation shows that the function values at the left and right ends of the interval are related by the linear transformation

$$\begin{bmatrix} \varphi(z_j) \\ \varphi'(z_j + 0) \end{bmatrix} = M_j \begin{bmatrix} \varphi(z_{j+1}) \\ \varphi'(z_{j+1} - 0) \end{bmatrix} \qquad (7)$$

where

$$M_j = \begin{bmatrix} \cosh \beta_j \delta_j & -\beta_j^{-1} \sinh \beta_j \delta_j \\ -\beta_j \sinh \beta_j \delta_j & \cosh \beta_j \delta_j \end{bmatrix}$$

and $\delta_j = z_{j+1} - z_j$.

Using (6) and (7) we determine the values $\varphi_i(0)$ and $\varphi_i'(0)$ from $\varphi_i(z_n)$ and $\varphi_i'(z_n)$. It seems reasonable to replace the boundary condition $\varphi_i(0) = 0$ by $\varphi_i(z_n) = 0$. For the K profiles

given in Figure 1 this is sufficient for Rn^{222} and its short-lived decay products, if $z_n = 3 \times 10$ cm, as in our case. However, the concentration of the long-lived Rn^{222} decay products is still not negligible at this altitude. For this reason the calculation was, in general, extended to the region $z_n = 30$ km $< z < \infty$ by putting $K =$ constant $= 3 \times 10^4$ cm²/sec for $z \geq z_n$. We then have $\varphi'(z_n)/\varphi(z_n) = -\beta_n$. Starting with an arbitrary initial value, $\varphi(z_n) = c$, and the initial value, $\varphi'(z_n) = -\beta_n c$, we computed the values $\varphi(z_j)$ and $\varphi'(z_j)$ for $j = n - 1, \cdots, 0$, successively. The boundary condition at z_0 was then satisfied by multiplication with a suitable factor.

To avoid floating-point overflow—the φ_j increase rapidly as z decreases—it was sufficient to choose c sufficiently small in most cases. The computer used (Siemens model 2002) admits values between 10^{-50} and 10^{50} for variable point computations. To include the less favorable cases we started with z_ρ instead of with z_n for which

$$\sum_1^{\rho-1} \alpha_i \delta_i < 100 \qquad \sum_1^{\rho} \alpha_i \delta_i \geq 100$$

This procedure is suggested by the exponential behavior of the solutions ($e^{100} \approx 10^{40}$). In fact, after satisfying the boundary conditions at $z = 0$, the values at z_ρ are negligible if $\rho < n$.

To estimate the error caused by replacing the K profile by a step function the computation was repeated in some cases with a larger number of steps. The deviations were negligible.

THEORETICAL RESULTS AND COMPARISON WITH EXPERIMENTAL DATA

Rn^{222}. Figure 2 shows the Rn^{222} profiles which were calculated with the typical K profiles given in Figure 1. They are standardized to a mean exhalation rate of 1 atom/cm² sec. Exhalation measurements at several places having normal Ra^{226} content of the soil material indicate an average exhalation rate of 0.2 to 1.5 atoms/cm² sec [Israel, 1962], in rather good agreement with the mean value of 1 atom/cm² sec which was estimated theoretically by Israel [1958, 1962] from the diffusion transport of Rn^{222} in the surface layer of the ground. It follows that the calculated Rn^{222} profiles in Figure 2 should be directly comparable with the results of measurements over continental areas having normal Ra^{226} content.
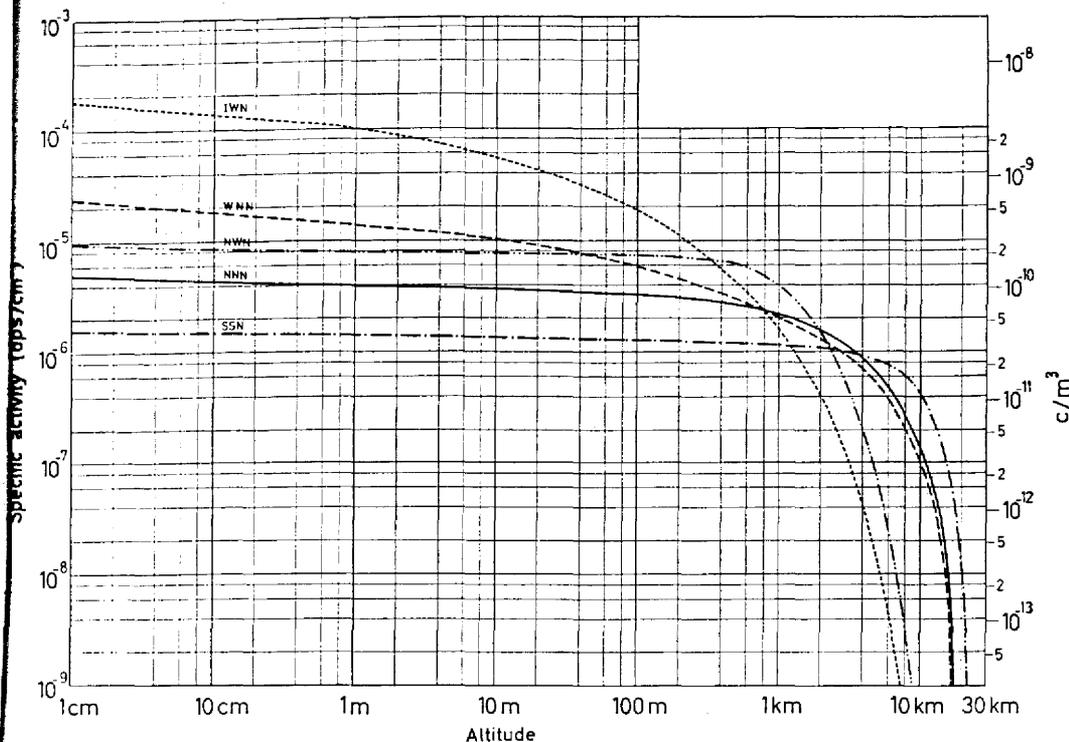
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Fig. 2. Vertical profiles of radon (Rn^{222}), calculated with $E_{Rn} = 1$ Rn atom/cm^2 sec for the K profiles *IWN, WNN, NWN, NNN*, and *SSN*.

Figure 2 indicates that the Rn^{222} concentration in the boundary layer, being strongly dependent on the mixing rate, varies by about a factor of 100 from a minimum value of about 0.3×10^{-10} c/m^3 for strong turbulence (case *SSN*) to a maximum value of about 30×10^{-10} c/m^3 for a strong inversion (case *IWN*). This range corresponds rather well to the observed fluctuations of the concentration near ground level. Compared with this variation, the influence of short time changes of the Rn^{222} exhalation from the ground is negligible. The theory indicates that a mean concentration of about 10^{-10} c/m^3 should be expected near ground level, in agreement with the mean values of most measurements at continental locations [*Hultqvist*, 1956; *Israel*, 1962].

Figure 2 also shows the interesting result that variations of turbulence have only a little influence on the concentration in the region from 0.5 to 2 km, because in this region the Rn^{222} profiles for different turbulence conditions are intersecting. For the rather extreme cases

which we have discussed, the concentration in this region varies only by about a factor of 5 as compared with a factor of 100 in the air near the earth's surface.

The radioactive decay of Rn^{222} ($t_{1/2} = 3.8$ d) leads to a monotonic decrease of the concentration with increasing altitude. According to the theory, a significant concentration gradient should be expected only in the boundary layer and for very weak turbulence, i.e., especially during strong inversion periods at night (case *IWN* in Figure 2). This theoretical conclusion can be checked by the observations of *Moses et al.* [1960]. Figure 3 shows observed Rn^{222} and temperature profiles at the end of nightly periods which had high stability and rather constant wind velocity. The observed profiles during strong inversions correspond rather well to the calculated profile for this case (profile *IWN*).

Beyond the ground layer over continental areas we made only a few series of Rn^{222} measurements which allow a comparison with the
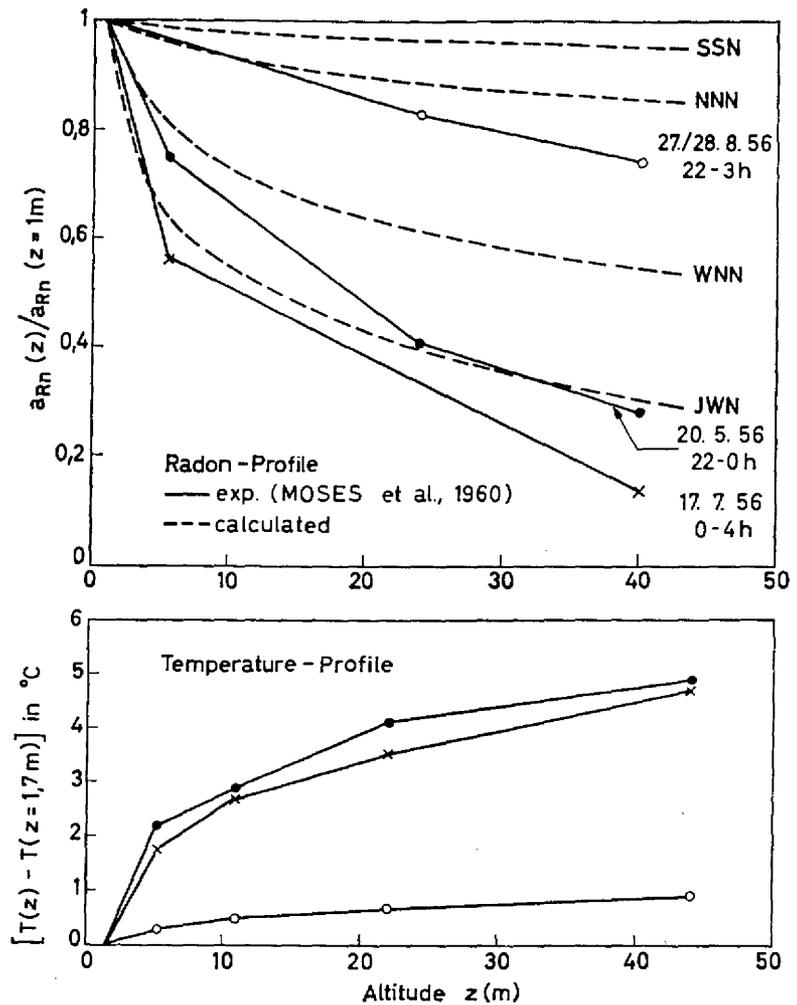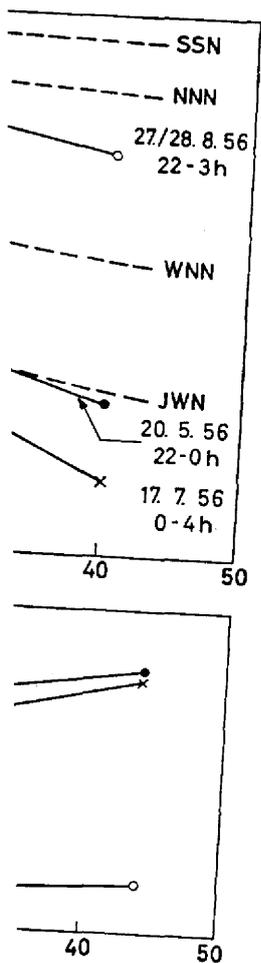
Fig. 3.   Comparison of observed and calculated Rn profiles near ground level under inversion
conditions.

theoretical results. Figure 4 shows two relative Rn[222] profiles which were observed by *Wexler et al.* [1955] during two flights over Ohio. They are compared with Rn[222] profiles which were calculated with equal exhalation rate from the K profiles *WNN, NNN,* and *SSN* (see Figures 1 and 2); the Rn[222] profile *NNN* was standardized to a concentration of 15 cpm/g at 2-km altitude. The profile observed on the first flight (Oct. 25, 1951) agrees rather well with the theoretical vertical distribution for the turbulence case *WNN.* The slope of the profile from the following day corresponds in the lower troposphere to the calculated Rn[222] profile for strong turbulence (case *SSN*) but approaches

the profile for the *NNN* case (normal turbulence) in the upper troposphere. The mean profile of both flights agrees rather well with the calculated profile *NNN* for which the Rn[222] concentration at an altitude of 2 km is about one-half the concentration near ground level. This is also consistent with observations of *Wigand and Wenk* [1928] and *Wilkening* [1953].

   This comparison indicates that it is possible to derive the vertical profile of the turbulent diffusion coefficient from measurements of the relative vertical Rn[222] distribution. The available measurements within the troposphere give some confidence that the K profile *NNN*, given in Figure 1, indeed represents average turbulence

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------ SSN

------ NNN

27/28. 8. 56
22-3h

------ WNN

~ JWN
20. 5. 56
22-0 h

17. 7. 56
0-4h

40   50

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Fig. 4. Comparison of observed and calculated relative Rn profiles in the troposphere.

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conditions in the troposphere. If we assume normal conditions of turbulence in the troposphere, i.e., the profile *NNN* in Figure 1, a rather rapid decrease of the Rn^{222} concentration should be expected in the tropopause. In this case the part of the total activity in a vertical column of air which reaches the layer above 12 to 13 km (mean altitude of the tropopause in the temperate zone) is only about 1 to 2 per cent. If we assume the K profile *SSN* (strong tropospheric turbulence), however, this part in-

creases to about 10 to 15 per cent. This suggests that considerable amounts of Rn^{222} can penetrate through the tropopause into the lower stratosphere only during periods of strong vertical turbulent mixing or convection, e.g. in case of cumulonimbus formation.

Machta and Lucas [1962] recently reported the first results of Rn^{222} measurements in the tropopause and lower stratosphere which indicate new aspects of the exchange process through the tropopause (Figure 5). The air

samples were taken in the polar region above Alaska (70°N) and in the equatorial region south of Hawaii (12° to 20°N). All three profiles of the Alaska series indicate a large negative gradient in the 8- to 12-km layer, i.e., in the tropopause. Above this layer in all cases the concentration again increases to a maximum at about 15 to 16 km and then decreases with increasing altitude.

For the interpretation of these results, two Rn^{222} profiles (A and B) are plotted in Figure 5. They were calculated for an exhalation rate of 1 atom/cm² sec and the corresponding K profiles A and B are given on the right side of the figure. In the tropopause the gradient of profile B is similar to the observed slope during the Alaska series. If equilibrium conditions are assumed, this slope indicates a mean turbulent diffusion coefficient of 1×10^4 to 3×10^4 cm²/sec in this air layer. In the Hawaii series the decrease in the tropopause region is less pronounced; it corresponds to an average diffusion coefficient of 5×10^4 to 1×10^5 cm²/sec. This high rate of turbulence is not consistent with the high vertical gradient of fission product and W^{185} activity in the lower equatorial stratosphere which was observed after the nuclear weapons tests in the equatorial stratosphere. It must be concluded that the high Rn^{222} content in the lower equatorial stratosphere is mainly due to upward-directed convection, which may occur especially above continental areas, rather than to turbulent diffusion.

The observed increase in concentration in the 12- to 15-km layer during the Alaska series cannot be explained by a steady-state equilibrium in an atmosphere which is horizontally isotropic. It may be explained either by horizontal advection of Rn^{222}-enriched air in the lower stratosphere, which overlaps the tropopause layer, or by nonequilibrium conditions of the vertical exchange process at the sampling location.

The first interpretation was given by *Machta and Lucas* [1962], who suggest that the Rn^{222} in the 15-km layer comes from the equatorial troposphere and enters the lower polar stratosphere through the tropopause gap. In this case the Rn^{222} in the 15-km layer above Alaska should have a more recent tropospheric history than that in the tropopause region below. Since the Rn^{222} content of this layer is about the same at polar and equatorial sampling sites, the transit time for the transport from south to north through the tropopause gap must be rather short, probably not exceeding a week.

Another interpretation of the observed profile over Alaska is possible if a sudden temporary decrease of vertical mixing or convection is assumed. If, for instance, there is a sudden change of the K profile from type A to type B (Figure 5), the supply of fresh Rn^{222} from the troposphere to the stratosphere is interrupted. Because of the low turbulence rate in this layer, most of the old Rn^{222} in the lower stratosphere remains there until it decays. A fraction of the old Rn^{222} in the tropopause will diffuse downward into the troposphere or upward into the lower stratosphere. Since the mixing rate is higher in the tropopause than in the lower stratosphere, the Rn^{222} content decreases more rapidly in the tropopause than in the layer above, and the result is a temporary, slow increase in concentration with altitude above the tropopause.

A check on possible contaminations of the air samples and further measurements are necessary to confirm these preliminary results. In any event they indicate the value of Rn^{222} as a tracer in the study of the exchange between troposphere and stratosphere. The main reason for its value is its rather short half-life of 3.8 days, which sets a time scale for the processes involved.

Short-lived Rn^{222} decay products. For the decay products Po^{218} ($t_{1/2} = 3.05$ min), Pb^{214} ($t_{1/2} = 26.8$ min), and Bi^{214} ($t_{1/2} = 19.7$ min), the theory predicts, as expected, radioactive equilibrium with Rn^{222} except in surface air. In the boundary layer the radioactive equilibrium is disturbed because of the deposition of decay products at the earth's surface which results in a downward diffusion flux within the boundary layer.

Figure 6 shows the vertical profiles of Po^{218}, Pb^{214}, and Bi^{214} (Po^{214}) in the boundary layer, which were calculated with the aid of the K profiles given in Figure 1. In all cases a lack of the three decay products with respect to Rn^{222} must be expected for steady-state conditions. This deficiency decreases with increasing height above ground level. At a given height the Pb^{214}/Rn^{222} and Bi^{214}/Rn^{222} ratios are nearly equal but are lower than the Po^{218}/Rn^{222} ratio.

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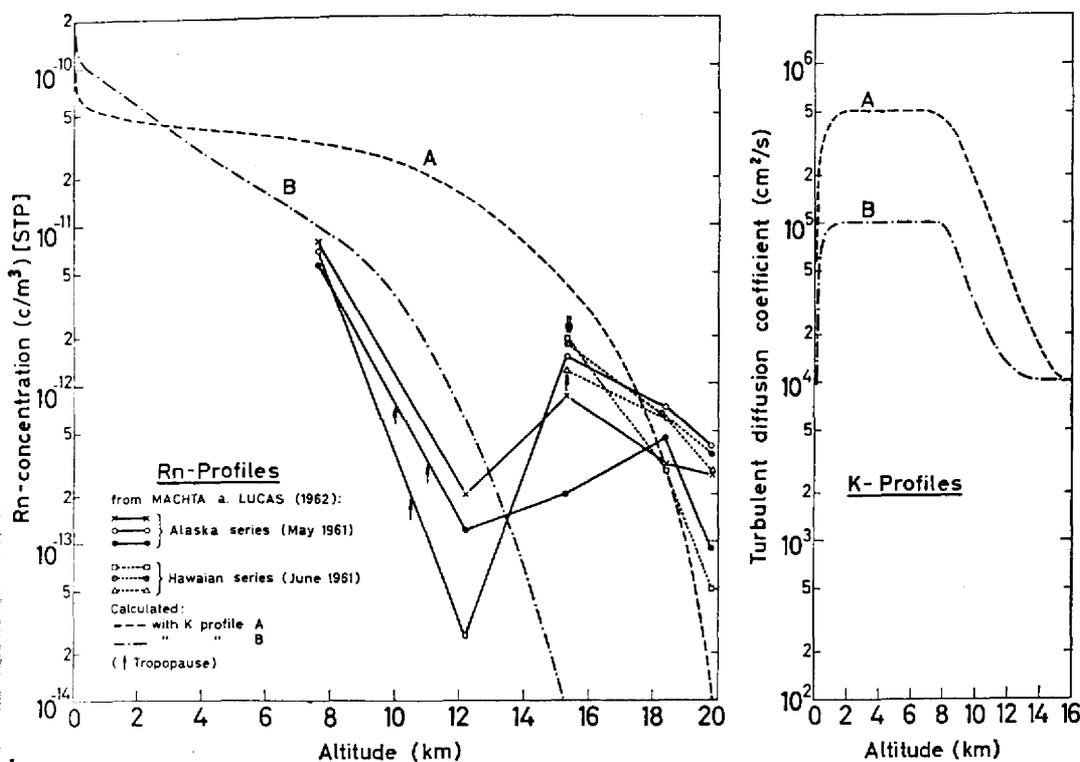)f the disturbed layer depends on
e within the boundary layer. For

Fig. 5. Comparison of observed Rn profiles in the upper troposphere and lower stratosphere over Hawaii and Alaska with theoretical profiles (A, B) calculated with the K profiles on the right figure.
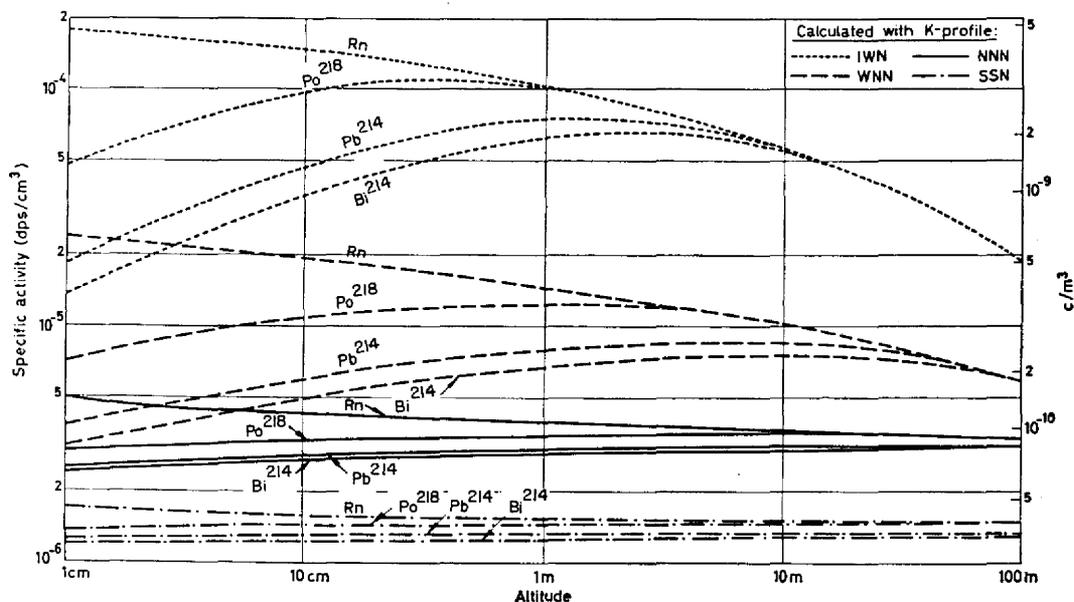

Fig. 6. Calculated vertical profiles of radon and its short-lived decay products in the boundary layer near ground level (E_{Rn} = 1 Rn atom/cm² sec).

TABLE 1. Calculated Deviation of Radioactive
Equilibrium between Rn²²² and Its Short-Lived
Decay Products

Nuclide Ratio	Turbu-lence Case	Activity Ratio			
		Height, m			
		0.1	1	10	100
Po²¹⁸/Rn²²²	IWN	0.66	0.98	1.00	1.00
	WNN	0.56	0.85	0.99	1.00
	NNN	0.77	0.88	0.95	1.00
	SSN	0.90	0.93	0.96	0.98
Pb²¹⁴/Rn²²²	IWN	0.31	0.73	0.99	1.00
	WNN	0.31	0.54	0.82	0.99
	NNN	0.66	0.76	0.84	0.94
	SSN	0.83	0.85	0.88	0.91
Bi²¹⁴/Rn²²²	IWN	0.23	0.62	0.97	1.00
	WNN	0.25	0.45	0.73	0.88
	NNN	0.63	0.72	0.80	0.90
	SSN	0.80	0.82	0.84	0.88

low turbulence (cases IWN and WNN) the de-
ficiency is rather large near ground level but
decreases rapidly with increasing height. With
increasing turbulence the deviation from radio-
active equilibrium with Rn²²² will be smal
near ground level but will extend to grea
heights (see Table 1).

An experimental proof of the disturbance
the boundary layer of the radioactive equil
rium between Rn²²² and Pb²¹⁴ or Bi²¹⁴ is possi
by simultaneous measurements of Rn²²² with
ion chamber or the scintillation chamber metl
and of Pb²¹⁴ and Bi²¹⁴ by the filter method.
few simultaneous measurements of this ki
though in a rather undefined terrain, were m
by *Jacobi et al.* [1959]. The average Bi²¹⁴/R
ratio for 35 measurements at a height of 1 to
meters—most of them during daytime—was
to 0.5; radioactive equilibrium was found c
during rather strong inversions at night anc
the morning. Minimum values of about 0.2
0.3 were observed in the noon and afternoon

An exact proof of the theoretical results
quires measurements of the Rn²²² and I
(Bi²¹⁴) profiles in the boundary layer during
riods of rather constant atmospheric stabil
But the theoretical results demonstrate the
sibility of obtaining valuable information on

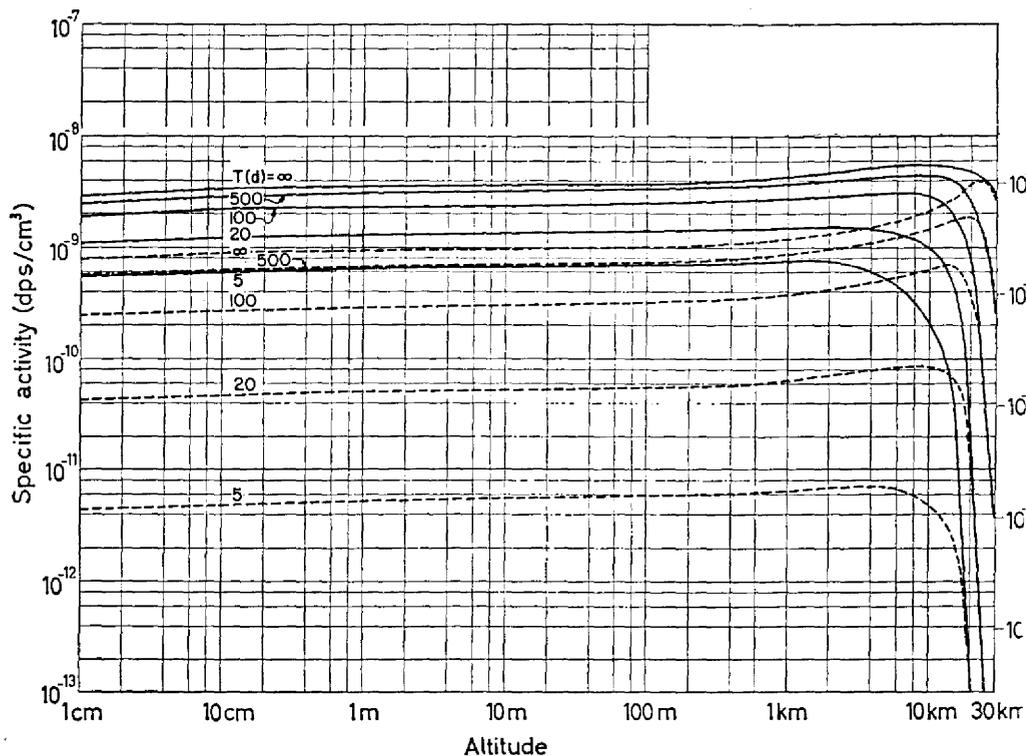Fig. 7. Vertical profiles of Pb²¹⁰ and Po²¹⁰, calculated with $E_{Rn} = 1$ Rn atom/cm² sec and with
the K profile NNN (parameter T = half-life of removal).

librium with Rn²²² will be smaller
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proof of the theoretical results re-
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btaining valuable information on the
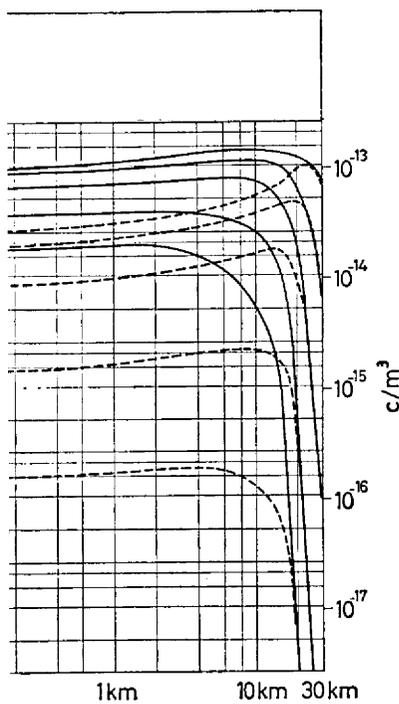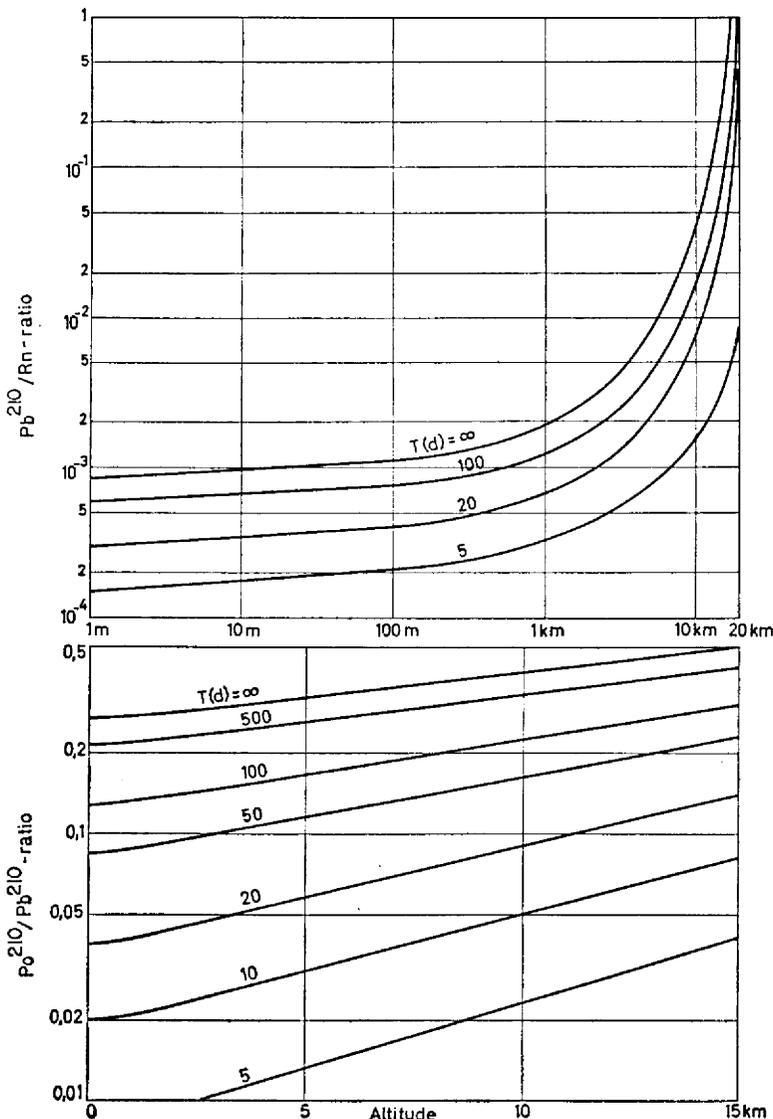

Rn = 1 Rn atom/cm² sec and with
.fe of removal).

Fig. 8. Vertical variation of the Pb²¹⁰/Rn and Po²¹⁰/Pb²¹⁰ ratios for different values of the removal half-life T (calculated with K profile NNN).

mixing rate in the boundary layer from mea-
surements of the ratios of Rn²²² and its decay
products.

Pb²¹⁰ and Po²¹⁰. The vertical distribution of
Pb²¹⁰ ($t_{1/2} = 19.4$ years) and Po²¹⁰ ($t_{1/2} = 138$ days) is limited by the washout and rainout of these nuclides in the troposphere. Their removal was taken into account by including the term with the removal rate Λ in (1) and (2). The corresponding half-life of elimination is

$T = (\ln 2)/\Lambda$. The assumption of a constant value of Λ is but a rough approximation, valid only for average conditions and time periods which are long compared with the duration of single precipitation processes. Because of the nature of this assumption, calculations of the equilibrium Pb²¹⁰ and Po²¹⁰ profiles were not justified for extreme turbulence conditions; therefore, the computations were restricted to the mean K profile NNN given in Figure 1.

TABLE 2. Calculated Mean Values of Pb^{210} and Po^{210} Concentrations and Their Ratio near Ground Level as a Function of the Removal Rate

Removal Half-Life T, days	Spec. Activity,* c/m³		Activity Ratio Pb^{210}/Po^{210}
	Pb^{210}	Po^{210}	
5	0.6×10^{-14}	0.5×10^{-14}	0.008
10	0.9×10^{-14}	1.8×10^{-16}	0.02
20	1.3×10^{-14}	5.0×10^{-16}	0.04
50	1.9×10^{-14}	1.6×10^{-15}	0.08
100	2.4×10^{-14}	2.9×10^{-15}	0.12
200	2.7×10^{-14}	4.6×10^{-15}	0.17
∞	3.5×10^{-14}	9.8×10^{-15}	0.28

* Assuming a mean Rn^{222} exhalation rate of 0.4 atom/cm² sec.

Figure 7 shows the calculated equilibrium profiles of Pb^{210} and Po^{210} for different values of the half-life of removal T. The profile of Bi^{210} ($t_{1/2} = 5.0$ days) is approximately identical with the Pb^{210} profile because both nuclides are practically in radioactive equilibrium. The distribution for $T = \infty$ applies to the case with no washout or rainout, so that fallout is effected only by diffusion to the ground surface. All the calculated profiles show a slow increase of the Pb^{210} and Po^{210} concentrations with altitude in the troposphere. We have to keep in mind that the calculations are based on the assumption of a constant removal rate throughout the troposphere and lower stratosphere. Since the precipitation processes are restricted to the troposphere, the actual profiles in the lower troposphere should correspond to profiles calculated for a finite residence time. Above the tropopause, however, a higher Pb^{210} and Po^{210} concentration should be expected. The maximum concentration should be reached at an altitude of about 20 to 30 km.

An indicator of the fallout efficiency is the change of the Pb^{210}/Rn^{222} and Po^{210}/Pb^{210} ratios with altitude. The theoretical values of these ratios are shown in Figure 8. The ratios are approximately constant up to an altitude of about 1 km. Above this altitude the Pb^{210}/Rn^{222} ratio increases rapidly according to the decrease of Rn^{222} concentration and exceeds a value of 1 in the lower stratosphere. The Po^{210}/Pb^{210} ratio rises more slowly with increasing height, until

radioactive equilibrium is reached approximat in the lower stratosphere.

The theory shows the important result tl rather low ratios must be expected in the trop sphere even without any removal of Pb^{210} a Po^{210} by washout or rainout. In this case Pb^{210}/Rn^{222} ratios of about 10^{-2} and Po^{210}/Pb^{210} rat of about 0.3 are obtained in ground-level a This result can be understood by considering t buildup of the steady-state distributions Pb^{210} and Po^{210} atoms. The vertical distributi of the production rate of Pb^{210} atoms must equal to the vertical profile of Bi^{214} or Rn^{222}. B cause of their comparatively long residence tir in the atmosphere, the Pb^{210} atoms assume much more uniform vertical distribution th; their mother atoms. Supported by the diffusic of Pb^{210} atoms toward the earth's surface ar their deposition thereon, this process results a lack of Pb^{210} in the troposphere and in an e: cess with respect to Rn^{222} in the lower strat(sphere. The deviation of radioactive equilibriur between Pb^{210} and Po^{210} without any washor. can be explained in the same way. With d(creasing residence time of Pb^{210} and Po^{210} atom i.e. with increasing washout efficiency, both r: tios are lowered.

The profiles of Pb^{210} and Po^{210} shown in Fig ure 7 are standardized to a Rn^{222} exhalation rat of 1 atom/cm² sec. As was mentioned earlie1 this value is reasonable for the calculation c Rn^{222} profiles over continental areas when th horizontal advection of maritime air with lov Rn^{222} content can be neglected. Because of thei rather long residence times in the atmosphere this is no longer true for Pb^{210} and Po^{210}. As ar approximation it can be assumed that horizonta: mixing leads to uniform Pb^{210} and Po^{210} profile: over land and sea within the same latitude belt. The land/sea ratio for middle latitudes of the northern hemisphere is about 0.4. The calculated absolute concentrations should be multiplied by this factor to get values that are comparable with the observed average concentration: in this region. Table 2 summarizes the resulting theoretical mean concentrations of Pb^{210} and Po^{210} near ground level for different value: of the half-lives of their removal.

Only a few measurements of Pb^{210} and Po^{210} are available for comparison with the calculations. Observed average values in ground-level air at several places in temperate latitudes of

quilibrium is reached approximately
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without any removal of Pb^{210} and
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rage values in ground-level
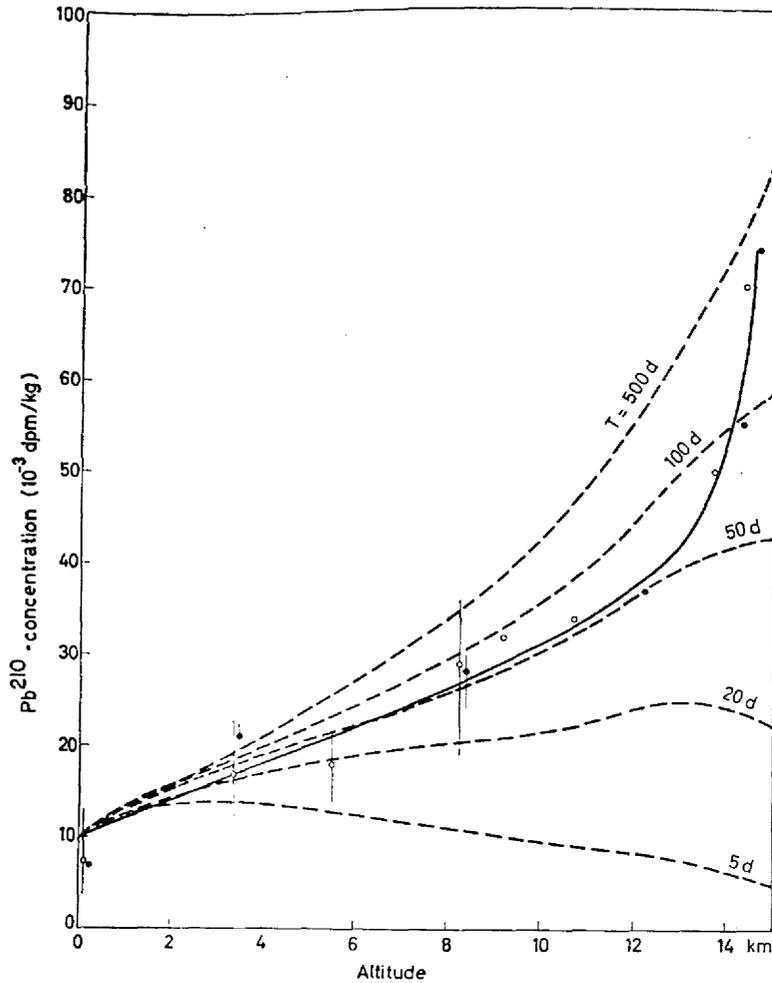in temperate latitudes of

Fig. 9.   Observed mean Pb^{210} profile over England, solid line, compared with the calculated Pb^{210} profiles for different removal half-life T (K profile NNN), dashed lines.

the northern hemisphere range from 0.5×10^{-14} to 2×10^{-14} c/m^3 for Pb^{210} [*Blifford et al.*, 1952; *Lehmann and Sittkus*, 1959; see also *US-AEC Fallout Program*, 1958, 1959, 1960]. Simultaneous measurements were made only by *Burton and Stewart* [1960] in Harwell and by *Lehmann and Sittkus* [1959] on the Schauinsland near Freiburg/Brsg. They obtained an average Po^{210} concentration of 5×10^{-14} to 7×10^{-14} c/m^3 and a mean ratio of 0.05 to 0.1. These experimental data are in rather good agreement with Table 2 for a removal half-life of 20 to 30 days. A half-life of this magnitude is well established by studies of the fallout of fission products [*Stewart*

et al., 1955] and cosmic-ray-induced Be^7 and P^{32} [*Goel et al.*, 1959; *Lal*, 1959].

Burton and Stewart [1960] measured the vertical distribution of Pb^{210} and Po^{210} over England during a period of several months. Figure 9 shows the observed mean Pb^{210} profile and the theoretical profiles (calculated for the density profile in the 'standard atmosphere'). The observed slow increase in concentration with altitude in the troposphere agrees rather well with the profile calculated with $T = 20$ to 50 days. Above the tropopause the concentration increases more rapidly and approaches the calculated profile for $T = \infty$. This effect should be
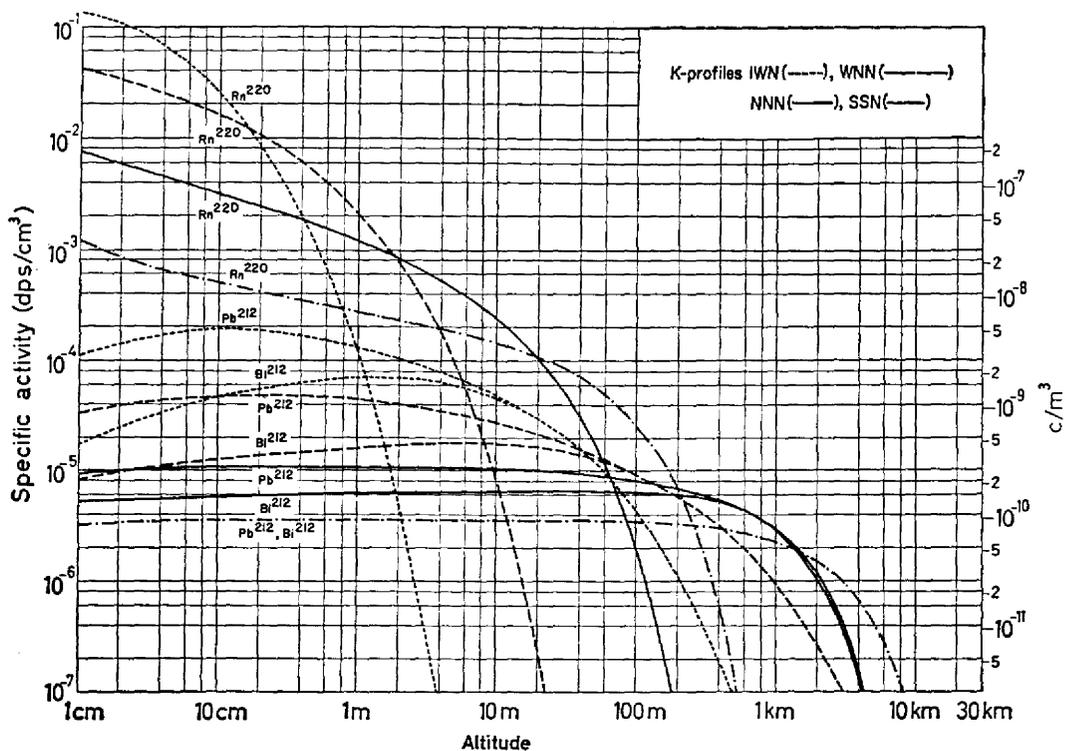
Fig. 10. Vertical profiles of Rn^{220} and its decay products, calculated with $E = 1$ atom/cm^2 sec.

expected from theory because of the lack of precipitation in the stratosphere. This agreement leads to the conclusion that the observed profile can be explained by vertical mixing without assuming a horizontal advection of Pb^{210}-enriched air in the lower stratosphere. Preliminary measurements of the profile over the United States, which were mentioned by *Machta* [1960], indicate no significant increase in the concentration above the tropopause. If this discrepancy is confirmed by further investigations, a dependence of the Pb^{210} profile on longitude must be assumed.

Burton and Stewart [1960] observed an increase of the Po^{210}/Pb^{210} ratio in air with increasing altitude as expected from theory. This is also consistent with the observation of *Lehmann and Sittkus* [1959] that the ratio in rain is higher by a factor of about 2 than that in ground-level air.

Rn^{220} and its decay products. Owing to the rather short half-life of Rn^{220} ($t_{1/2} = 54$ sec) and its decay products, their vertical distribution is restricted to the lower troposphere. Figure 10

shows the vertical profiles of Rn^{220}, Pb^{212}, and Bi^{212} calculated for the typical profiles of the turbulent diffusion coefficient shown in Figure 1. Because of its short half-life, the concentration of Rn^{220} decreases rapidly with height. In the case *NNN* (average turbulence) about 80 per cent of all Rn^{220} atoms will decay within 20 meters above ground level. For a strong inversion in the boundary layer (case *IWN*) about 90 per cent of Rn^{220} is concentrated in a layer 1 meter thick. Po^{216} rather quickly approaches radioactive equilibrium with Rn^{220}.

The half-life of Pb^{212} ($t_{1/2} = 10.6$ hr) greatly exceeds that of Rn^{220}. Therefore, the Pb^{212} atoms, after their formation, can diffuse to greater altitudes than Rn^{220} or Po^{216}. As in the relations between Rn^{222} and Pb^{210}, this difference in residence time causes a low Pb^{212}/Rn^{220} ratio in the boundary layer near ground level and an excess of Pb^{212} over Rn^{220} at higher altitudes. Radioactive equilibrium between the two is reached at only one point, the altitude of which varies with mixing rate in the range from 1 to 100 meters. For a constant turbulent mixing rate the theory in-

les IWN(----), WNN(------)
NNN(----), SSN(----)

c/m^3

1km   10km 30km
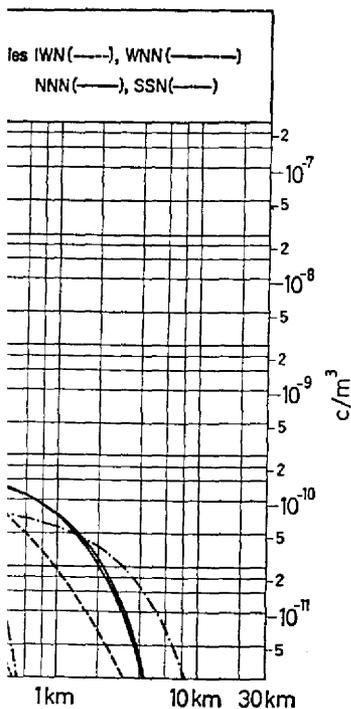
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n^{220} is concentrated in a layer
^{216} rather quickly approaches
orium with Rn^{220}.

Pb^{212} ($t_{1/2} = 10.6$ hr) greatly
^{220}. Therefore, the Pb^{212} atoms,
on, can diffuse to greater alti-
r Po^{216}. As in the relations be-
$)^{210}$, this difference in residence
Pb^{212}/Rn^{220} ratio in the bound-
ound level and an excess of
higher altitudes. Radioactive
n the two is reached at only
ude of which varies with mix-
ge from 1 to 100 meters. For
it mixing rate the theory in-

dicates a rather constant Pb^{214} concentration in
the range of 1 to 20 meters. For a constant Rn^{220}
exhalation, changes of the turbulent mixing rate
will cause a variation of the Pb^{214} concentration
in this layer by a factor of about 10.

The calculated profiles shown in Figure 10 are
standardized to a Rn^{220} exhalation rate of 1
atom/cm² sec. Measurements of the exhalation
rate are not available, but on the basis of the
diffusion theory and the known Rn^{222} exhalation
rate, a rough estimation is possible. Since the
average Ra^{226} and Th^{232} contents of normal soil
material are about equal and amount to 1.10^{-12}
c/g [*Rankama and Sahama*, 1950], the Rn^{220}
exhalation rate must be lower by a factor of
about $\sqrt{\lambda_{222}/\lambda_{220}} \approx 1/80$ than the Rn^{222} exhala-
tion rate, if both emanations escape from the
soil material with the same efficiency into the
ground air [*Israel*, 1958]. Consequently, a Rn^{220}
exhalation rate of 10^{-2} atom/cm² sec will be a
rough mean value for uncovered, dry ground of
normal Th^{232} content.

For comparison with natural conditions, the
vertical profiles shown in Figure 10 must there-
fore be lowered by a factor of about 0.01. For
the mean turbulence profile *NNN* the theory
predicts a mean Pb^{212} concentration of about
$3 \cdot 10^{-12}$ c/m³ and a mean Pb^{212}/Rn^{222} ratio of
about 0.03 in surface air (see Figure 10). Mea-
surements in this layer at several places [see
compilation of *Hultqvist*, 1956] indicate a mean
Pb^{212} concentration of 10^{-12} to 10^{-11} c/m³ and a
mean ratio of 0.01 to 0.05, which is in rather
good agreement with the theoretical values. The
assumed K profile *NNN* and the mean Rn^{220}
exhalation rate thus seem to be reasonable.

It follows from Figure 10 that it is impossible
to determine Rn^{220} concentration from Pb^{212} mea-
surements under the assumption of radioactive
equilibrium. Since no direct methods for the de-
termination of low Rn^{220} concentrations in the
presence of Rn^{222} are yet available, the Rn^{220} con-
centration in the atmosphere is not known. The
agreement between theory and observation,
stated above, for Pb^{212} allows us to conclude
that the calculated mean Rn^{220} profile for the
case *NNN* is quite correct under normal condi-
tions. Therefore, a mean Rn^{220} concentration of
about 10^{-10} c/m³ should be expected near ground
level, which is of the same order of magnitude
as the mean Rn^{222} concentration in this layer.

Conclusion. The theoretical calculations give

a general survey of the vertical distribution of
Rn^{222}, Rn^{220}, and their decay products in the at-
mosphere, and their dependence on the vertical
profile of turbulent mixing. The vertical dis-
tributions calculated for the average turbulence
profile *NNN* agree rather well with the ob-
served distribution of natural radioactivity in
the atmosphere. On this basis a reasonable pre-
diction of the Rn^{220} concentration in ground-
level air and its variation with altitude can be
given. On the basis of the theoretical results pre-
sented here it is necessary to revise the pre-
viously used method of estimating the mean
residence time of aerosols in the troposphere
from Pb^{210}/Rn^{222} or Po^{210}/Pb^{210} ratios.

The computed profiles show new aspects for
a successful use of Rn^{222}, Rn^{220}, and their decay
products as natural tracers in the study of mix-
ing processes in the troposphere and lower
stratosphere.

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