Investigations on radioactive equilibrium in the lower atmosphere between radon and its short-lived decay products

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ABSTRACT

The variations in the degree of equilibrium between radon and its short-lived decay products have been studied at 2 meters above the ground surface at a selected location near Trombay, Bombay, India. The correlations between the degree of equilibrium and different meteorological parameters such as atmospheric stability, wind speed and direction, atmospheric pressure, temperature, relative humidity and rainfall, have been studied. Diurnal and seasonal variations in the concentration of radon and in the degree of equilibrium have also been studied. The degree of equilibrium has been found to range from 4 to 100 % depending on meteorological conditions. This has been studied by making simultaneous measurements of actual radon decay product activity in a sample of air and the equilibrium decay product activity in the same air. The degree of equilibrium has been defined as the percentage of equilibrium daughter product activity actually present in a sample of air. The equilibrium daughter product activity has been measured by filtering out the daughter products from a known volume of air and subsequently allowing the daughter products to grow to equilibrium concentration in the presence of aerosols in a reservoir. The application of these investigations in understanding the mixing processes in the lower atmosphere have been discussed.

Introduction

A study of the dependence of the extent of radioactive equilibrium between radon and its short-lived daughters on various meteorological parameters would be highly useful for understanding small scale dispersion near the ground level. This paper describes the variations with time of radon concentration in the air and the extent of radioactive equilibrium at two meters above the ground level. The relationships of meteorological parameters such as atmospheric stability, range of temperature during a day, relative humidity, wind speed and direction, atmospheric pressure and rainfall with the radon content of the air and the extent of equilibrium have been studied by calculating the Pearson's co-efficient of correlation. This study shows that the measurements of radioactive equilibrium between radon and its short-lived daughter products can be advantageously made use of in understanding the diffusion process in the lower atmosphere.

Measurements

The measurements were made at Shantinivas in Mandala Village, 6 km north-east of the South Site of Bhabha Atomic Research Centre. This site is sparsely populated and has no tall buildings. However, there is bushy vegetation around the sampling location consisting of trees, mainly sapotaceae, varying from 2 to 5 meters in height. The measurements were made under natural conditions, without disturbing the soil cover.

A 5 meter tall pole was erected at the sampling site. The pole supported the ends of two polythene tubes at 2 meters above the ground level. One of these tubes led the air with radon to the apparatus used for determining the equilibrium daughter product activity and hence the radon concentration. The other tube connected a filter holder to a pump that sucked the atmospheric air through the filter paper for the measurement of actual daughter product activity. The samples of radon and its daughters from equal volumes of air were collected simultaneously.

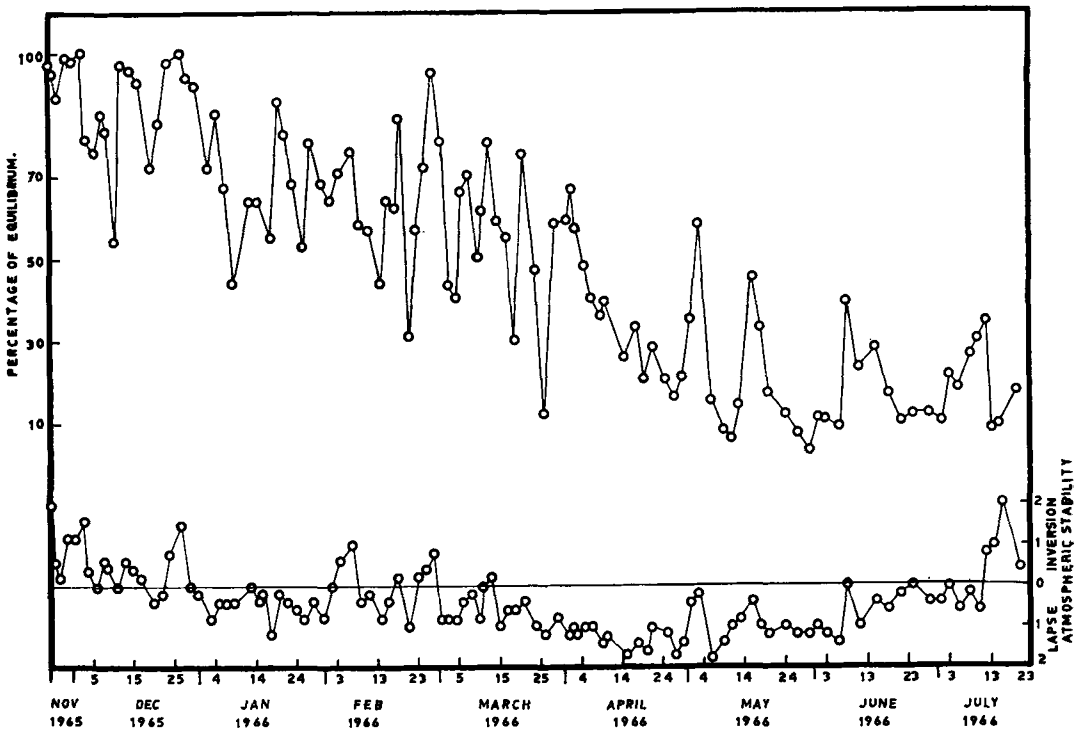

Fig. 1. Variation of percentage of equilibrium with atmospheric stability.

The method of determination of equilibrium daughter product activity was based on an earlier study which showed that when highly filtered and dried air mixed with submicron aerosol was allowed to stay for 3 hours in a reservoir, the radon in the air decayed and attained equilibrium with its short-lived daughter products which attached to the aerosol. The details of the method have been described in an earlier paper (Vohra, Subba Ramu & Mohan Rao, 1966). The samples were filled in the reservoir for periods of 30 minutes, along with the aerosol and after the stagnation time of 3 hours, the radon daughters attached to aerosols were collected on a millipore filter paper and their activity measured using an alpha scintillation counter. The short-lived radon daughters collected are Po^{218} (3.05 min), Pb^{214} (26.8 min) and Bi^{214} (19.7 min) in equilibrium with Po^{214}. Po^{218} and Po^{214} are alpha emitters. Since the sample is counted after 10 minutes, the alpha activity will be mostly due to Po^{214}.

The actual activity of radon daughter products in the free atmosphere was measured by drawing a known volume of air through the millipore filter paper. The activity collected on the filter paper was measured in the same way as above. The ratio of the actual radon daughter product activity to the equilibrium daughter product activity multiplied by 100 gave percentage equilibrium values.

Along with the measurement of radon con-

Table 1. *Monthly averages for radon concentration in the air and the percentage of equilibrium*

Month		Radon concentration in $\mu\mu c/m^3$	The percentage of equilibrium
December	1965	236.4	88.0
January	1966	224.0	68.0
February	1966	199.0	64.0
March	1966	165.5	55.5
April	1966	147.5	36.4
May	1966	139.8	21.3
June	1966	110.3	17.4
July	1966	147.6	19.4

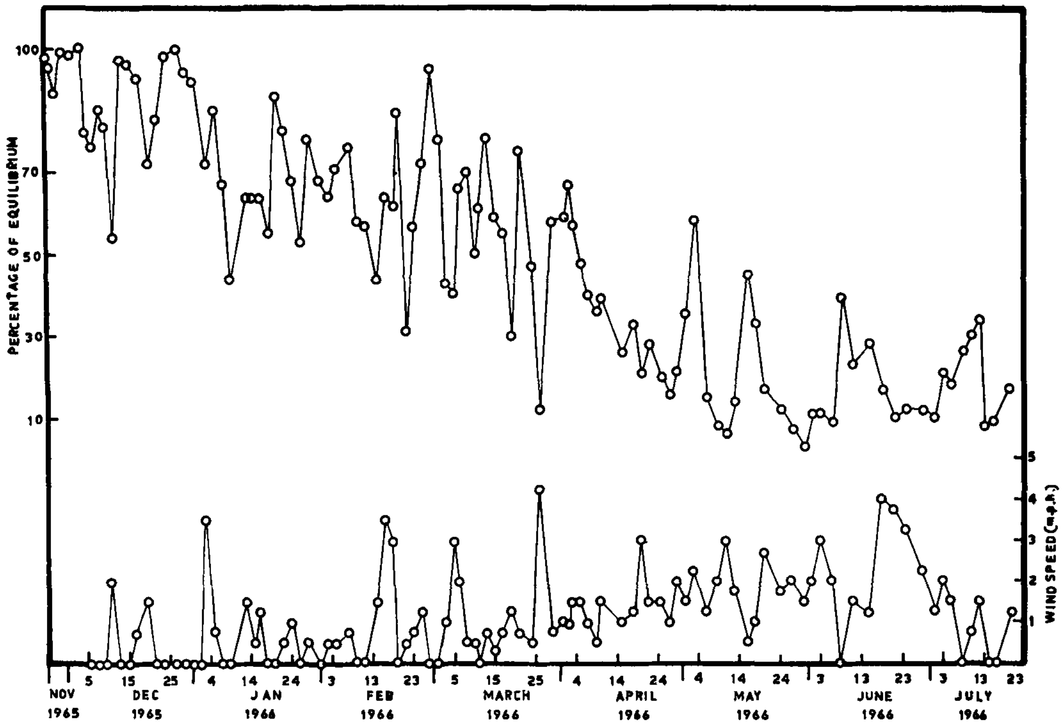

Fig. 2. Variation of percentage of equilibrium with wind speed.

centration and percentage of equilibrium the following meteorological parameters were also measured. The atmospheric stability was measured as the difference of temperatures between the heights of two and four meters. The temperature was measured using thermistors. The 5 meter tall pole was used for supporting the thermistors which were housed in louvered cone shades. The self-heating effect in the thermistors was overcome by using very small currents.

Table 2. *Two-hourly variations of radon concentration in the air, the percentage of equilibrium and meteorological parameters for the period from 1600 January 6, 1966 to 1600 January 7, 1966 at Shantinivas*

Sl. No.	Date	Time in hours	Radon concentration in $\mu\mu c/m^3$	The percentage of equilibrium	Temperature in °C: 25 cm	2 m	4 m	Range of temp.	Relative humidity	Wind: Speed (mph)	Direction
1	6.1.66	1615	207.3	81.2	28.6	29.8	31.4	11.0	42.0	Calm	
2	6.1.66	1815	215.8	76.2	21.0	21.6	23.6	11.0	67.0	Calm	
3	6.1.66	2015	278.6	73.0	—	—	21.6	11.0	79.0	Calm	
4	6.1.66	2215	304.9	70.0	—	—	—	11.0	83.0	Calm	
5	7.1.66	0015	315.3	71.0	—	—	—	11.0	84.0	Calm	
6	7.1.66	0215	314.4	89.4	—	—	—	11.0	82.0	Calm	
7	7.1.66	0415	347.9	66.0	—	—	—	11.0	84.0	Calm	
8	7.1.66	0615	366.7	76.1	—	—	—	11.0	84.0	Calm	
9	7.1.66	0815	363.7	87.1	—	—	—	11.0	88.0	Calm	
10	7.1.66	1015	331.3	75.2	24.4	26.4	25.8	11.0	52.0	Calm	
11	7.1.66	1215	239.1	67.4	31.0	31.4	31.2	11.0	34.0	Calm	
12	7.1.66	1415	244.5	58.6	32.0	33.6	33.6	11.0	25.0	Calm	
13	7.1.66	1615	236.5	60.8	29.0	30.2	31.0	11.0	32.0	1.00	NW

Table 3. *Two-hourly variations of radon concentration in the air, the percentage of equilibrium and the meteorological parameters for the period from 1000 February 11, 1966 to 0800 February 12, 1966 at Shantinivas*

Sl. No.	Date	Time in hours	Radon concentration in $\mu\mu c/m^3$	The percentage of equilibrium	Temperature in °C 2 m	Temperature in °C 4 m	Range of temp.	Relative humidity	Wind Speed (mph)	Wind Direction
1	11.2.66	1015	262.4	56.7	28.2	28.0	15	57	Calm	
2	11.2.66	1215	253.0	36.1	34.0	33.6	15	40	Calm	
3	11.2.66	1415	134.4	78.3	31.2	31.0	15	50	1.0	NW
4	11.2.66	1615	163.9	55.7	30.6	31.4	15	54	1.0	NW
5	11.2.66	1815	167.0	64.3	27.6	28.6	15	66	1.5	NW
6	11.2.66	2015	323.8	40.1	25.2	25.2	15	93	Calm	
7	11.2.66	2215	335.0	37.1	22.2	22.2	15	93	Calm	
8	12.2.66	0015	374.4	37.8	21.0	21.4	15	100	Calm	
9	12.2.66	0215	295.6	46.5	20.0	20.0	15	100	Calm	
10	12.2.66	0415	340.6	40.9	20.0	20.0	15	97	Calm	
11	12.2.66	0615	295.6	52.7	20.0	20.0	15	95	Calm	
12	12.2.66	0815	378.8	43.0	21.2	21.4	14	95	Calm	

Severn readings of temperature differences between 2 and 4 meters were taken and averaged over the sampling period.

The range of temperature during a day was measured by a maximum–minimum thermometer. The relative humidity was measured by wet and dry bulb thermometers. Average of four sets of readings of dry and wet bulb temperatures taken during the period of sampling were calculated and the relative humidity corresponding to the atmospheric pressure was read from a standard table.

An anemometer with a range of 0 to 70 m.p.h. was used for wind speed measurements. When the wind speed was less than 0.5 m.p.h., it was noted as "calm". Four sets of wind speed measurements were made during the sampling period and the mean wind speed was calculated. Wind directions along with the wind speed measurements were noted and the predominant wind direction was taken into consideration. An aneroid barograph with four chambers was employed for the measurements of atmospheric pressure. A natural siphon rain recorder was used to measure the rainfall in millimeters.

The measurements of radon concentration

Table 4. *Two-hourly variations of radon concentration in the air, the percentage of equilibrium and the meteorological parameters for the period from 1000 March 26, 1966 to 0800 March 27, 1966 at Shantinivas*

Sl. No.	Date	Time in hours	Radon concentration in $\mu\mu c/m^3$	The percentage of equilibrium	Temperature in °C 2 m	Temperature in °C 4 m	Range of temp.	Relative humidity	Wind Speed (mph)	Wind Direction
1	26.3.66	1020	131.2	18.1	28.6	28.6	11.5	87	1.0	NW
2	26.3.66	1215	99.9	12.1	31.2	30.2	11.5	92	4.3	NW
3	26.3.66	1415	108.8	12.3	31.2	30.0	11.5	85	3.0	NW
4	26.3.66	1615	96.3	20.9	30.0	29.4	11.5	98	2.0	NW
5	26.3.66	1815	161.2	17.2	25.6	26.0	11.5	91	2.0	NW
6	26.3.66	2015	161.2	21.2	23.8	25.2	11.5	90	Calm	
7	26.3.66	2215	185.4	24.2	20.2	23.4	11.5	100	Calm	
8	27.3.66	0015	296.9	43.7	20.0	22.0	11.5	100	Calm	
9	27.3.66	0215	194.4	59.9	20.0	23.0	11.5	100	Calm	
10	27.3.66	0415	179.6	50.4	20.0	22.2	11.5	95	Calm	
11	27.3.66	0615	179.2	54.5	20.0	21.6	11.5	100	Calm	
12	27.3.66	0815	138.9	55.8	23.6	24.6	10.5	81	0.5	NW

Table 5. *Two-hourly variations of radon concentration in the air, the percentage of equilibrium and the meteorological parameters for the period from 1000 May 1, 1966 to 0800 May 2, 1966 at Shantinivas*

Sl. No.	Date	Time in hours	Radon concentration in $\mu\mu c/m^3$	The percentage of equilibrium	Temperature in °C 25 cm	2 m	4 m	Range of temp.	Relative humidity	Wind Speed (mph)	Direction
1	1.5.66	1015	161.2	42.2	34.0	35.0	33.8		31.0	1.25	NE
2	1.5.66	1215	179.6	34.9	36.8	36.4	36.0	13.0	44.0	1.50	N
3	1.5.66	1415	206.0	35.4	37.4	36.8	36.0		38.0	2.25	N
4	1.5.66	1615	271.0	47.6	35.0	35.4	34.6		42.0	4.25	NW
5	1.5.66	1815	268.7	51.7	33.0	33.0	33.0		46.0	1.00	NW
6	1.5.66	2015	300.9	58.0	29.8	30.6	30.6		60.0	Calm	
7	1.5.66	2215	291.9	53.7	28.4	28.8	30.0		52.0	Calm	
8	2.5.66	0015	282.1	55.7	29.2	29.8	30.4		57.0	Calm	
9	2.5.66	0215	300.0	86.6	28.2	28.4	29.0		75.0	Calm	
10	2.5.66	0415	354.6	65.0	26.8	26.8	27.8		82.0	Calm	
11	2.5.66	0615	389.5	44.8	28.0	28.0	28.4		79.0	1.00	E
12	2.5.66	0815	322.4	40.9	30.6	30.8	30.8	10.0	66.0	1.00	E

and degree of equilibrium were made on alternate days during the period November 1965 to July 1966. Sampling time was 30 minutes and all measurements were made between 1100 and 1200 hours. Diurnal variations of the radon content of the air and the extent of equilibrium were studied on the following four days.

1600 hrs of 6.1.1966 to 1600 hrs of 7.1.1966
1000 hrs of 11.2.1966 to 0800 hrs of 12.2.1966
1000 hrs of 26.3.1966 to 0800 hrs of 27.3.1966
and 1000 hrs of 1.5.1966 to 0800 hrs of 2.5.1966

Fig. 1 gives the variations in percentage equilibrium and atmospheric stability for the period November 1965 to July 1966. Monthly averages of radon concentration and percentage equilibrium are given in Table 1. Fig. 2 gives the variations of percentage equilibrium with wind speed.

Diurnal variations of radon concentration, percentage equilibrium, atmospheric stability and wind speed on the above four typical days are given in tables 2 to 5. Fig. 3 gives a graph showing variations of radon concentration in the air with atmospheric pressure for the period 1.5.66 to 2.5.66. Fig. 4 gives the variations in radon concentration and percentage equilibrium during the periods of rainfall.

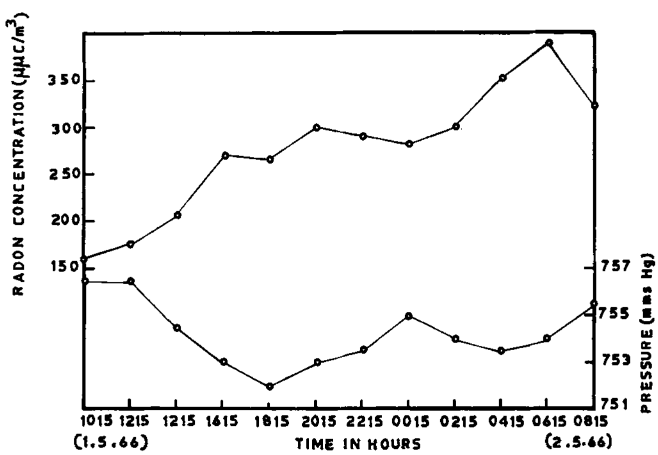

Fig. 3. Variation of radon concentration in the air with atmospheric pressure.

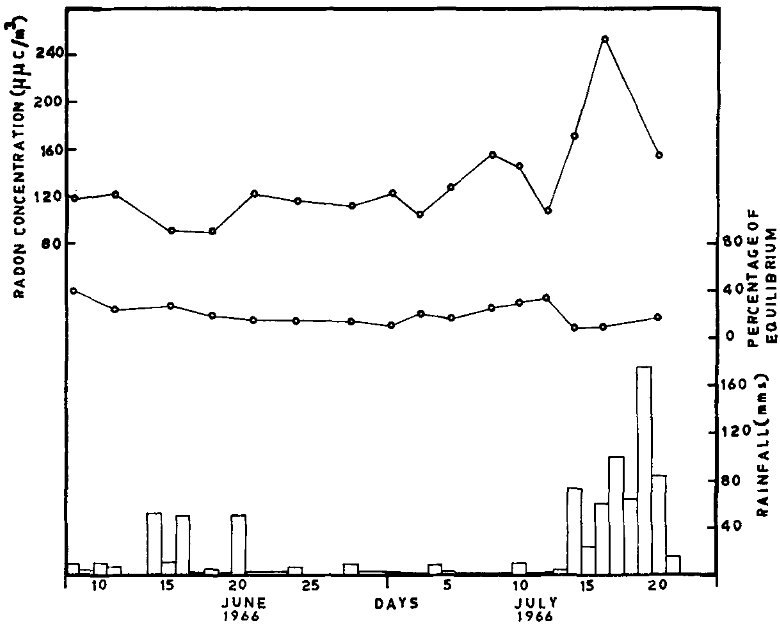

Fig. 4. The effect of rainfall on radon concentration and the percentage of equilibrium in the air.

Distribution of radon and its daughters in the lower atmosphere

Before we discuss the above results we shall consider the factors affecting the vertical distribution of radon and its daughters. The distribution of any of the radionuclides in the decay chain of radon and its change with time at a point in the atmosphere are given by

$$\frac{\partial n_i}{\partial t} = \nabla(K\nabla n_i) - U\nabla n_i + V\frac{\partial n_i}{\partial t} + n_{i-1}\lambda_{i-1} - n_i(\lambda_i + \Lambda_i) \quad (1)$$

where i denotes the position of the nuclide in the decay chain, n the number of atoms, U_x, U_y, U_z, the components of mean wind velocity, K, the turbulent diffusion coefficient, V, the mean sedimentation velocity, λ, the radioactive decay constant and Λ, the mean removal rate of the nuclide by rain out or wash out.

Let us consider an area from which the amount of radon gas escaping per unit area is constant. For steady-state conditions we can write $\partial n_i/\partial t = 0$. Also $(\partial n_i/\partial x) = (\partial n_i/\partial y) = 0$ for a horizontal isotropic distribution.

The steady-state condition may not hold good for large changes in atmospheric pressure and during rainfall. Significant changes in the radon content of the air at ground level have been observed for large changes in atmospheric pressure and during the period of rainfall. (Figs. 3 and 4). An inverse correlation between radon concentration in the air and the atmospheric pressure (when the deviation from the normal is large) measured for the period May 1, 1966 to May 2, 1966, has been found (Fig. 3) and 37 % of variance in one is associated with variation in the other.

But for these changes, the rate of injection of radon into the atmosphere from the soil may be considered fairly uniform. Israel (1951) observed that the exhalation rate of radon from soil to air is relatively constant over a known period and for a given area. Pearson (1965) has found that unless anomalous conditions such as the passage of a strong frontal system occur during a sampling period, temporal variation in emanation rate from soil may be neglected.

Further, $U_z = 0$ since in most of the cases the vertical wind velocity will be small compared with the velocity of K_z and $V_i \approx 0$ since the mean radioactivity radius for short-lived radon decay products in the atmosphere near the ground is around 0.09 micron (Jacobi *et al.*, 1959) and the corresponding sedimentation velocity is smaller than about 0.05 inch per hour, which is very small compared to the transport velocity due to turbulent mixing. $\Lambda_i = 0$ or negligible since for short-lived radon decay products $\lambda_i \gg \Lambda_i$. Our findings on the effect of rainfall on the concentration of radon and its daughter products in the atmosphere agree with those of Malakhov and Solodikhina (1962) who have found that the reduction in the concentration of radon and its short-lived decay products during the falling precipitation is insignificant. However, radon concentration near the ground level decreases as a result of reduction of exhalation due to clogging of soil capillaries.

With the above conditions and limitations, we obtain the following differential equations for radon and its short-lived decay products from equation (1)

$$\frac{d}{dz}\left(K\frac{dn_1}{dz}\right) - n_1\lambda_1 = 0 \quad \text{for radon} \qquad (2)$$

and

$$\frac{d}{dz}\left(K\frac{dn_i}{dz}\right) + n_{i-1}\lambda_{i-1} - n_i\lambda_i = 0 \qquad (3)$$

for short-lived decay products.

The boundary conditions for the above equations are

(*a*) $\int_0^\infty n\lambda dz = E$ where E is the exhalation rate of radon from the ground surface. Also $E_i = 0$ since no decay product of radon escapes from ground to the air.

(*b*) $n_i(z = 0) = 0$ for radon decay products. The radon decay products that reach the ground by diffusion are all deposited.

(*c*) $n_i(z \to \infty) \to 0$, due to radioactive decay.

K is quite variable with altitude according to the vertical variation of wind velocity and atmospheric stability. In the boundary layer near the earth's surface K_z increases rapidly following an approximately linear or power law of Z. The value of K_z in the atmosphere varies from 10 to 10^4 cm^2/sec at 1 meter and 10^4 to 10^6 cm^2/sec at 1 km above the ground level, depending upon the meteorological conditions.

Jacobi & André (1963) have calculated the vertical profiles of radon and its short-lived daughters in the lower layers of the atmosphere for four typical K_z profiles using equations (2) and (3). The four K_z profiles are (*a*) weak diffusion which is typical of strong inversion near the ground, (*b*) rather weak vertical mixing throughout the lower atmosphere, (*c*) normal turbulence conditions and (*d*) strong vertical mixing in the atmosphere. The exhalation rate of radon has been assumed to be $E = 1$ atom/cm^2/sec. Their calculations show that non-equilibrium between radon and its decay products may be expected in the lower atmosphere, decreasing with increasing height. This height depends on the mixing rate within the lower layers of the atmosphere. For low turbulence the non-equilibrium is rather large near ground level but decreases rapidly with increasing height. With increasing turbulence the deviation from radioactive equilibrium is smaller but will extend upto greater heights.

From the data in Fig. 1, it has been found that the association between the percentage of equilibrium and the atmsopheric stability is only 49% during the periods of temperature inversion whereas it is 84% during lapse. Also the range of temperature during a day (not shown in the figure) does not show any correlation with the percentage of equilibrium during inversion. This indeed reveals that the part played by forced convection is important in determining the diffusion co-efficient and suggests that the degree of equilibrium is a very sensitive measure of mixing rate of air masses in the lower atmosphere.

During lapse, the range of temperature during a day shows 64% association and the atmospheric stability 84% association with the percentage of equilibrium. This can be possible if the non-equilibrium between radon and its daughter products extends upto greater heights in the atmosphere and if the effect of vertical wind profile on the atmospheric diffusion is not predominant. Lapse in the atmosphere normally indicates a thorough mixing in which case a greater degree of equilibrium at 2 meters above the ground level should be expected. But on the other hand, observations show a decrease in this location. Also a positive correlation exists between the radon content of the air and the percentage of equilibrium (Table 1) irrespective of whether there is temperature inversion or lapse in the atmosphere.

It has been found that the degree of equilibrium decreases when the wind speed increases to more than 2 m.p.h. But, only a small frequency of higher wind speed has been observed throughout the period of investigation (Fig. 2) and the maximum wind speed observed is 4 m.p.h. only. These observations suggest that turbulence in the location selected for the measurements is rather weak even during temperature lapse.

The two hourly measurements of radon and the percentage of equilibrium during 24-hour intervals show a decided periodic variation. Night time values of radon concentration are usually higher than the day-time values, the maximum occurring in the early morning hours when inversion is known to occur or the lapse-rate is small (Table 3). During this period of increase of radon concentration, a decrease in the percentage of equilibrium is observed as expected for weak diffusion. The diurnal variations observed are as given in Tables 2 to 5. In almost all the cases of the diurnal variations, wind speed was less than 0.5 to 1 m.p.h. during the nights. However, even the small scale variations in wind speed have an effect on the equilibrium values. For example in the month of March, 1966 (Table 4) the radon concentration decreased while the percentage of equilibrium increased in spite of inversion. Variations in the diffusion co-efficient K_z due to those in the vertical gradient of wind speed affect the radon concentration and the percentage of equilibrium in the atmsphere. This variation, even if it is on a small scale, can be quite significant during inversion since according to Jacobi & Andrè, the disequilibrium between radon and its daughter products varies rapidly with height for weak diffusion.

Now we shall consider the experiments conducted by Richardson (1925) on the nature of turbulence and vertical temperature differences near trees. The criterion for eddies to vanish (just-no-turbulence) is

$$\left(\frac{\partial V_x}{\partial Z}\right)^2 + \left(\frac{\partial V_y}{\partial Z}\right)^2 = \frac{g}{T}\left(\frac{\partial T}{\partial Z} - \frac{\partial T_A}{\partial Z}\right)$$

where g is the acceleration due to gravity and
$\partial T/\partial Z$ is the up-gradient of temperature,
$\partial T_A/\partial Z$ is the adiabatic up-gradient of temperature,
$\partial V/\partial Z$ is the up-gradient of wind speed.

Richardson made observations of temperature and wind profiles upto 26 meters in a location with trees of non-uniform size, kind and arrangement. He found that statistically unstable vertical gradients of temperature were common between heights of 4.4 and 18.3 meters in the middle of the day. To compare the observations with the above formula a diagram was drawn having vertical temperature differences and vertical wind differences as the co-ordinates. The curve represented by an equation obtained using the above formula was plotted on the diagram and was found that it did really lie near the boundary of the region of no gusts. Most of the observations were found to lie far away from the criterion in the gusty region. The measurements of the extent of equilibrium between radon and its short-lived daughter products also show that turbulence is rather low near trees.

Summary and Conclusions

(1) Non-equilibrium exists between radon and its short-lived decay products in the lower atmosphere. The percentage of equilibrium can vary between the limits of 4 and 100, depending upon the meteorological conditions.

(2) Decided periodic variations are found for a 24 hour interval in the radon content of air and the extent of equilibrium. Within an hour or two the variations are not significant except during the periods of transition from night to day and vice versa. This can be explained on the basis of diurnal variation in the intensity of atmospheric turbulence.

(3) Seasonal variations of radon concentration and the percentage of equilibrium are observed. The maximum and the minimum radon concentration in the air are 318 $\mu\mu c/m^3$ and 90 $\mu\mu c/m^3$ respectively. For the maximum value the extent of equilibrium was 92% and for the minimum it was only 18% for the location studied. Higher levels of radon content of air and the percentage of equilibrium are found in the winter season and a gradual decrease is found towards summer and monsoon seasons.

(4) Initial rainfall after a long period of dryness causes an increase in the radon content of air at 2 meters above the ground level. Subsequent rainfall however causes plugging effect and the escape of radon from soil to air is retarded. Also rain has only a slight filtering power for radon daughters in the atmosphere.

(5) The degree of equilibrium at 2 meters above the ground level during inversions corresponds to that estimated by the computations (by Jacobi & André) of the vertical distribution of radon and its decay products for the K_z profile corresponding to weak diffusion. During

lapse conditions the percentage of equilibrium decreases, contrary to the expectations, since the local vegetation has an influence on the vertical diffusion in the lower atmosphere.

(6) The nature of turbulence at the location chosen for these investigations as studied from the measurements of the percentage of equilibrium is same as that observed by Richardson whose experiments have shown that turbulence near trees is rather weak.

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ИССЛЕДОВАНИЕ РАДИОАКТИВНОГО РАВНОВЕСИЯ В НИЖНЕЙ АТМОСФЕРЕ МЕЖДУ РАДОНОМ И КОРОТКОЖИВУЩИМИ ПРОДУКТАМИ ЕГО РАСПАДА

На определенных площадках вблизи Тромбея, Бомбей, Индия, на высоте 2 *м* изучались вариации степени равновесия между радоном и короткоживущими продуктами его распада. Изучались корреляции между степенью равновесия и различными метеорологическими параметрами, такими, как стратификация атмосферы, направление и скорость ветра, давление, температура, относительная влажность и дожди. Изучались также суточные и сезонные вариации концентрации радона и степени радиоактивного равновесия. Было найдено, что степень равновесия в зависимости от метеорологических условий меняется от 4 до 100%. Это было получено путем одновременных измерений действительной активности продуктов распада радона в пробе воздуха и активности равновесных продуктов распада в той же самой пробе. Степень равновесия определялась как процент активности равновесных дочерних продуктов, присутствующих в данной пробе. Активность равновесных дочерних продуктов измерялась путем отфильтровывания дочерних продуктов из известного объема воздуха и последующим предоставлением возможности дочерним продуктам возрасти до равновесных концентраций в резервуаре в присутствии аэрозоля. Обсуждается применение этих исслодований к изучению процессов перемешивания в нижней атмосфере.