Simultaneous measurements of radon (Rn^{222}) and thoron (Rn^{220}) in the atmospheric surface layer

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ABSTRACT

Simultaneous, direct measurements of the radon and thoron contents in the air near the ground have been made, using apparatus adapted from those of Norinder et al. (1952) and Israel (1964). Data of the diurnal variations and the vertical distributions of radon and thoron are presented. According to measurements the vertical gradients of radon and thoron are greatest near the ground surface at night. The ratios between the concentrations of thoron and radon vary on an average from 0.1 to about 3.0 in the layer up to the height 3 m above the ground surface. Fourier analyses are made of variations with time of radon and thoron concentrations. For radon a few harmonics represent the diurnal wave. For thoron, however, several harmonics are needed.

Introduction

Radon and thoron are produced in the soil and transported into the atmosphere by turbulent transfer (Israel, 1962). There have been numerous researches into the relation between radon and thoron concentrations and the turbulent diffusiveness of the air (Schmidt, 1926; Jacobi & Andre, 1963; Crozier & Biles, 1966; Fontan et al., 1966; Israel et al., 1966; Malakhov et al., 1966; Hosler, 1969; Druilhet et al., 1971; Ikebe & Shimo, 1972; Cohen et al., 1972 and others).

The diurnal variations of radon concentrations in the atmospheric boundary layer are extensively documented.

Studies of the diurnal variations of thoron concentrations are, however, more scarce (Fontan et al., 1966; Israel, 1966; Druilhet et al., 1971 and others).

Direct measurements of the thoron concentrations in the atmosphere were impossible until some years ago (Fontan et al., 1962*a*, 1962*b*; Israel, 1964). Measurements of the thoron content in soil air are presented several years ago (Norinder et al., 1952).

The concentrations of radon, thoron and their decay products vary differently according to the time since their formation is due to different decay constants. Their use as tracers for studies of certain problems of atmospheric physics increases if radon, thoron and their decay products are measured simultaneously.

After 1969 radon, thoron and their decay products were measured in the atmospheric surface layer at Marsta Observatory. In previous papers, Israelsson et al. (1972*a*), (1972*b*) the natural α- and β-activity of the air near the ground are treated. The present paper deals with simultaneous measurements of the radon and thoron concentrations. The apparatus used here is ionization chambers with special electronic equipments.

The present study is part of a program for investigating the atmospheric electricity and radioactivity in the atmospheric boundary layer at Marsta Observatory.

Method of measuring the concentrations of radon and thoron

The concentration of radon and its daughter products is generally measured after having been concentrated by absorption or filtering from a fixed volume of air. Due to difficulties of measuring the thoron content of the atmosphere in general the thoron content is computed from

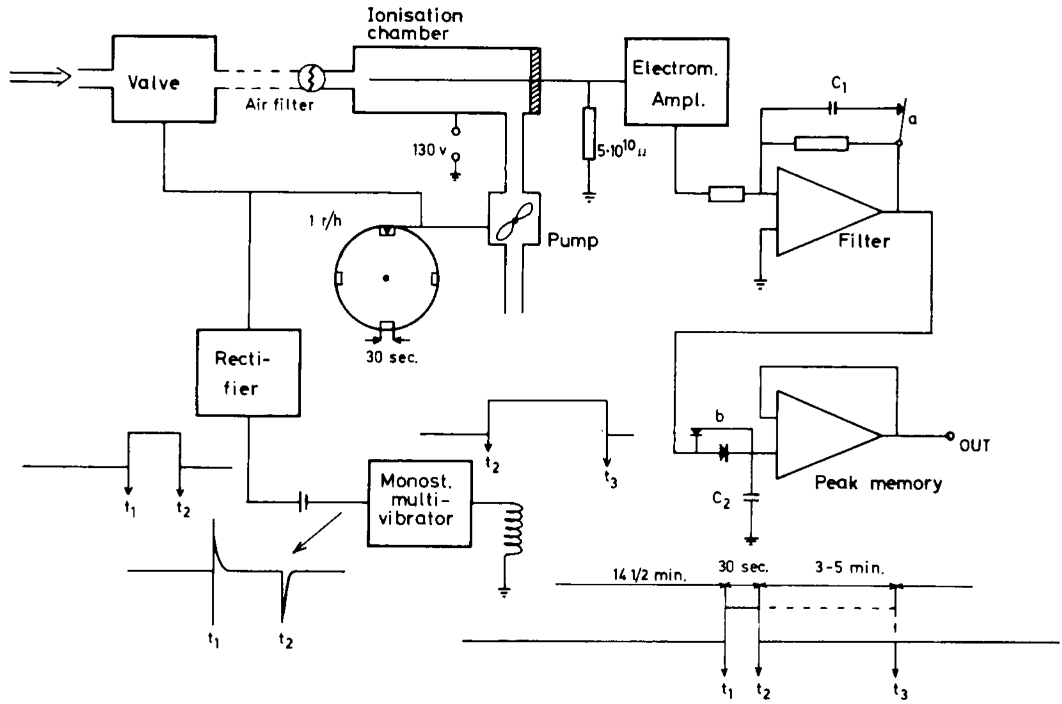

Fig. 1. Arrangement of the equipment for measuring of radon and thoron.

the content of ThB assuming equilibrium between Tn and ThB.

Direct measurements of the radon and thoron concentrations in the soil air are made by Norinder et al. (1952). A direct ionmetirc method was used for measuring the content of emanation.

Fontan et al. (1962*a*, 1962*b*) developed a method to measure the thoron concentration in the atmosphere, using an indirect method of enrichment, i.e. by collecting the ThB set free by the decay of thoron in a known volume of air.

In order to measure radon and thoron concentrations in the atmosphere continuously with a high time resolution aspirating ionization chambers were used (Israel, 1964, 1966).

Later the method described by Israel (1964) was used by Crozier & Biles (1966), Ikebe & Shimo (1972), Israelsson et al. (1972*a*) and others.

Measurements of natural radioactivity (Norinder et al., 1952) give for Uppsala a radon content some 10 times the normal values generally given in the literature. A high radon content in Uppsala makes it possible to make simultaneous measurements of both radon and thoron in the air near the ground. The method used by us is in principle according to methods described by Norinder et al. (1952), Israel (1964) and Israel & Israel (1966). The measuring method used by us allowing the recording of radon and thoron contents of the atmosphere as well as the radon and thoron contents in the soil air. From the radon profiles below the ground surface the exhalation rates of radon can be determined.

The mathematical theories for the ionization processes are given by Evans (1955). When radon and thoron are introduced into an ionization chamber the disintegration of thoron causes a rapid decrease of the ionization current. After that we obtain an increase in ionization current due to the accumulation of RaA and RaC′. Theoretical considerations are given by Israel (1957).

The ionization curve obtained after 10 min can be extrapolated back to the starting point using the theoretic accumulation curve for radon. Then the difference between this

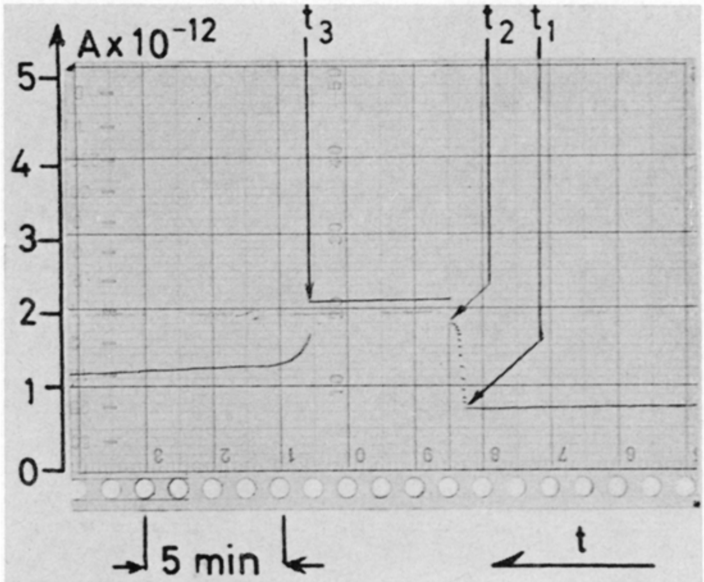

Fig. 2. Time expanded recording where the times t_1, t_2 and t_3 refer to the given description of the equipment in Fig. 1.

ionization current and the total ionization current gives the ionization current for thoron (see Norinder et al., 1952).

Some practical problems will arise from the recording of radon and thoron, viz.:

(1) the ionization current will fluctuate and an electrical filter is therefore required

(2) a time constant gives an unreal picture of the decay curve of thoron. Consequently during the initial time the smoothening filter ought to be compensated for by a very low time-constant.

These problems are here solved with a special electronic equipment.

Fig. 1 shows the arrangement of the equipment. The measurement units consist of a 24 liter ionization chamber and an electrometer amplifier (Analog Devices 311). The sensitivity of the electrometer is for the accuracy $\pm 1\%$ about 10^{-13} Amp. The other units include time-constants and units for programming the recording.

The timer starts the air pump at the time t_1. The valve is opened and atmospheric air is sucked through the air filter into the ionization chamber. The function of the air filter is to prevent radioactive aerosols from entering the chamber. The valve prevents an air exchange through the chamber during the measuring.

Further, a cylinder condenser with a high electrical field is placed between the air filter and the chamber. Small ions are, thus, prevented from entering the chamber.

After about 30 sec at the time t_2, the air pump stops and the valve is closed. At the time t_2 a monostable multivibrator is triffered on the negative pulse (shown in the figure) and a relay on the out-put of the multivibrator is operating at about 3 to 5 min and the contacts *a* and *b* are opened. The filter amplifier is now working as a unit gain amplifier as fast as C_1 is disconnected. The filter function is consequently out of use. On the other hand, a peak memory is connected, viz. the peak out-put voltage of the filter amplifier is stored on C_2 during a great part of the decay time of thoron.

After about 3 to 5 min (variable), at the time t_3, the relay contacts *a* and *b* are closed and the filter function is connected while the peak-reader function is disconnected. The recording represents now the radon and its decay products. The voltage curve is now smoothened due to the connected condenser. Fig. 2 shows a time expanded recording where the times t_1, t_2 and t_3 refer to the given description of the equipment.

A typical recording with a recording speed of 20 mm/hour is shown in Fig. 3. The individual

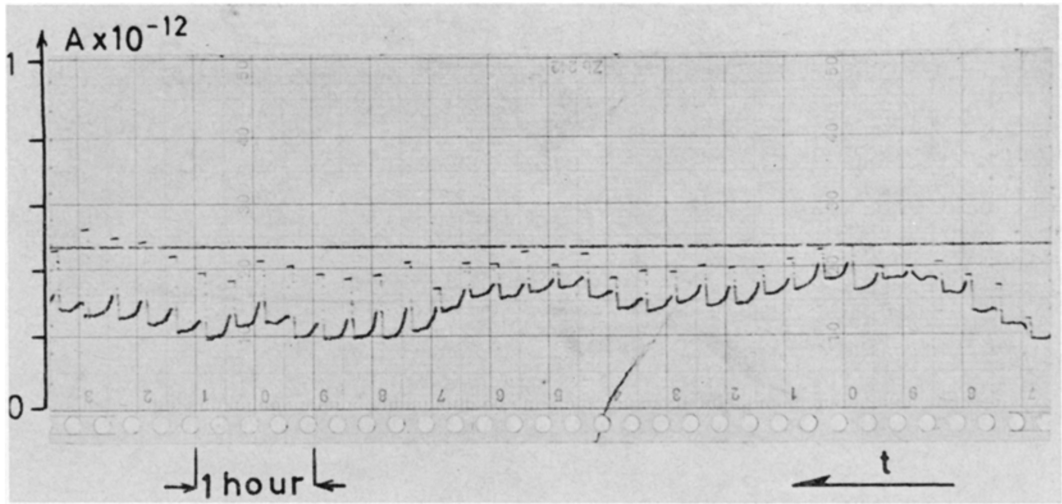

Fig. 3. Recording of ion current in the apparatus. The individual measurements are repeated every 15 min.

measurements are repeated every 15 min. Radon and thoron can now be separated and determined if the background radiation is known.

Calculation of the thoron and radon content

For calibration the ionization chamber is tested with radon gas. The radon activity is measured at Statens Strålskyddsinstitut, Stockholm, with an accuracy of $\pm 6\%$. In the calibrations the ionization current is determined against the radon activity. From the calibrations, the detection limit is found to be approximately 0.1 pC/l.

According to the theory of the ionization processes in an ionization chamber the radon curve can be extrapolated back to the starting point. The differences between the ionization current at this point and the 'peak-value' give us the value of thoron concentration. The accuracy of the thoron measurements is limited by the statistical fluctuations of the background noise plus the radon decay, whilst the accuracy of the radon measurements depends on the background fluctuations. The background radiation is remarkably constant, so it could be compensated for almost completely. On an average the accuracy is about 10% over much of our range of measurement.

The decay of thoron during the passage through the intake tube

Measurements of the profiles of radon and thoron in the layer near the ground require a smoothed area with no obstacles, for instance buildings. The ionization chamber therefore is placed at some distance from the point of measurement. In the intake tube the thoron content decreases due to the radioactive decay. This decay is of less importance for radon.

Theoretically, this correction can easily be made when the air speed in the tube is known.

Representativeness of the gradient measurements of radon and thoron due to the intake tube

Because of the short half-life time of thoron (54.4 sec) rapid measurements have to be made. Therefore large air-flows are required. These circumstances disturb the natural condition near the ground where the radon and the thoron gradients are significant (see Crozier & Biles, 1966).

In order to obtain correct profiles near the ground different types of intake tubes are used, see Fig. 4. The left tube has only one big hole and the right one has 70 small holes distributed over 3 m. We tested the two types of intake tubes, by measuring the radioactivity at three heights with the two intake tubes. Fig. 5 illustrates the differences in using the two intake tubes. The ordinate represents the height above the ground surface and the abscissa refers to the different values using the two types of intake tubes. In order to show the difference between the two intake tubes the

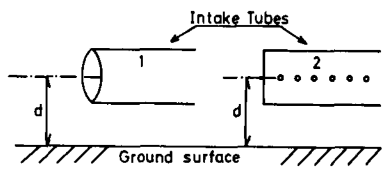

Fig. 4. Different types of intake tubes. The left tube has only 1 hole and the right one has 70 small holes distributed over 3 m.

ratio $(R_{70} - R_1)/R_1 \cdot 100$ is calculated. R_1 = activity value when only 1 hole is used and R_{70} = activity value when 70 holes are used.

The diagram represents measurements under conditions with near-neutral and light unstable stratifications. The horizontal lines represent the standard deviation from 14 measurements.

In the diagram we can note that both types of intake tubes can be used at heights 15 cm and higher above the ground surface. But near the ground surface we had to use a tube consisting of several holes which seems to permit greater certainty as to the height from which the air is being drawn.

The thoron profiles are often determined near the ground surface. Therefore it should be favourable near the ground surface to suck the air from larger areas instead of only one point. In the measurements near the ground surface we therefore use an intake tube with a length of 3 m and consisting of 70 holes. At other heights we use tubes with only 1 hole.

Variations of radon and thoron concentrations at ground level

In studies of different matters in the atmospherie knowledge of the distribution of the matters both in space and with time is of great importance. The present section deals with studies of the time variations of radon and thoron. The vertical distribution of these gases are also treated.

The distribution of the radioactive gases radon and thoron in the air can be given by (see i.e. Wilkening, 1970)

$$\frac{\partial C}{\partial t} = \nabla(K \cdot \nabla C) - \underline{v} \cdot \nabla C - \lambda C \qquad (1)$$

where C = concentration of radon or thoron, K = coefficient of eddy diffusivity, v = mean wind and λ = decay constant of radon or thoron.

Eq. (1) with simplifying assumptions have been treated, among others, by Jacobi & Andre (1963); Staley (1966); Fontan et al. (1966); Israel (1966); Hosler (1969); Druilhet et al. (1971); Cohen et al. (1972); Ikebe & Shimo (1972) and others.

Vertical distributions of radon and thoron

In the present study we will treat simultaneous measurements of the vertical distributions of radon and thoron at Marsta Observatory.

There have been numerous researches into the relation between radon and thoron concentrations and the distance above the ground surface. There have, however, been a limited number of simultaneous measurements of the vertical distributions of radon and thoron concentrations (see i.e. Jacobi & Andre, 1963; Fontan et al., 1966).

To obtain representations of radon and thoron activity against distance above the ground surface, we made simultaneous measurements of radon and thoron at different heights. The radon and thoron concentrations are determined every quarter of an hour. The measurements are made in June–August 1971.

Figs. 6 and 7 represent the vertical distribution of radon and thoron respectively.

Profiles A in Figs. 6 and 7 apply to daytime conditions in sunny weather. Profiles B apply to night-time conditions in situations with stable stratifications.

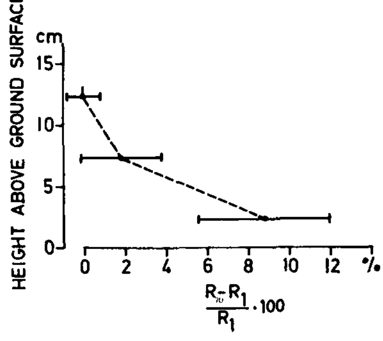

Fig. 5. The ratio $(R_{70}-R_1)/R_1 \cdot 100$ at different heights above the ground. R_1, Activity value when 1 hole is used; R_{70}, activity value when 70 holes are used.

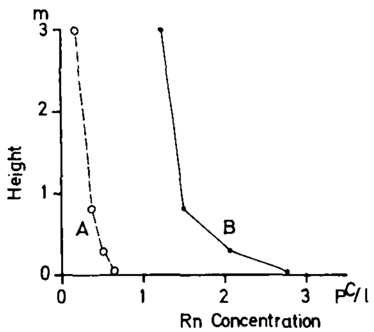

Fig. 6. Vertical distribution of radon concentration. *A*, sunny days; *B*, night-time.

In the diagrams in Figs. 6 and 7 we observe the influence of the turbulent transfer upon the radon and thoron profiles. In the daytime the turbulent transfer is large which causes small gradients of radon and thoron. At night the turbulent transfer is small and the vertical gradients of the pnatural radioactive gases are therefore great.

The half-life times of the two gases (radon 3.8 days and thoron only 54.5 seconds) cause the remarkable differences of the gradients.

In Fig. 7 we also observe at the height of about 1 m above the ground small differences in thoron activity between night and day. For radon, however, great concentrations are observed at all heights at night.

The profiles of thoron seem to be fairly similar to those obtained by Crozier & Biles (1966) and Ikebe & Shimo (1972). Further radon profiles similar to those in Figs. 6 and 7 are observed by several researches.

From the diagrams we note that the ratio between the radon and thoron concentrations is quite near the ground surface 2.9 in daytime and 1.6 at night. At the height 3 m above the ground surface the ratios are 0.9 in daytime and 0.1 at night.

In a later paper we will present exchange coefficients of radon, thoron and momentum obtained from profile measurements of radon, thoron, wind speed and temperature in the atmospheric surface layer.

Time variations of the radon and thoron concentrations

The variations of radon and thoron in the surface layers depend mainly upon the following two factors: the exhalation rates of radon and thoron from the ground surface and the turbulent transfer of radioactivity from the surface layer to the upper layers of the atmosphere.

The intensity of the turbulent transfer depends to a very high degree upon the wind speed and the vertical stability of the air both of which vary according to the meteorological situation—during the day and the seasons of the year.

In the present investigation we treat the diurnal variation of radon and thoron at the height 0.3 m above the ground. Fourier analyses of the time variations of radon and thoron in the surface layer are also made.

Simultaneous measurements of the diurnal variations of radon and thoron

The diurnal variation of natural radioactivity in the air near the ground is mainly determined by the diurnal variation of the turbulent transfer. This is treated in several investigations, see i.e. Wilkening (1959), Fontan et al. (1966), Israelsson et al. (1968) and others.

The effect of the variation of the exhalation rate of radon and thoron is theoretical considered by Malakhov et al. (1966).

In the present study the diurnal variations of radon and thoron at the height 0.3 m above the ground are studied during fine weather days in May–June 1970. Hourly-means are computed from 14 days. The mean-values are then plotted in Fig. 8.

In the diagrams we note a diurnal variation of the two gases. Radon shows the most pronounced variation with about 0.6 pC/l in daytime and between 4 and 5 pC/l at night. Thoron concentration is about 2 pC/l in the daytime and 6 pC/l at night.

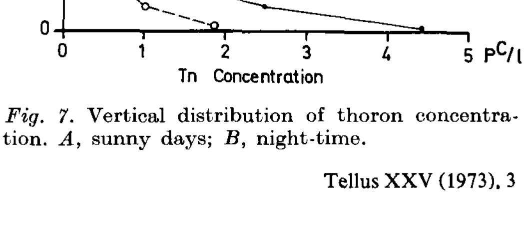

Fig. 7. Vertical distribution of thoron concentration. *A*, sunny days; *B*, night-time.

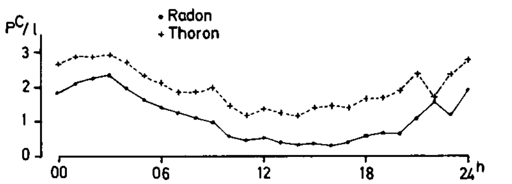

Fig. 8. Diurnal variations of radon and thoron concentrations at the height 0.3 m.

The great turbulent transfer in the daytime causes small radon and thoron concentrations in the daytime and a small turbulent transfer at night seems to cause great concentrations of natural radioactivity at night.

The intensity of the turbulent transfer of natural radioactivity depends to a very high degree upon the wind and temperature conditions both of which vary according to the meteorological situation, see i.e. Wilkening (1959); Reiter (1964) and others. We have therefore calculated the mean diurnal variation of the wind speed at the height, 10 m, u, and the temperature stratification, here expressed by the temperature difference between the levels 32 m and 1.5 m above the ground. The data are plotted in Fig. 9. The measuring period is the same as in Fig. 8.

From the diagrams in Figs. 8 and 9 it is apparent that the concentrations of radon and thoron are very dependent on the wind speed and the temperature stratification. Maximum values of the radioactivity occur for the lowest wind speed and greatest the lapse-rate of the temperature, viz. during surface inversions.

The smallest activity values are noted when wind is greatest and the lapse-rate of the temperature is negative, viz. under conditions with unstable stratification in the daytime.

The diurnal variation of the radon concentration seems to be similar to those obtained by Fontan et al. (1966); Pearson & Moses (1966) and others.

For the thoron concentrations, however, different measurement sites influence fairly much upon the time variations of the thoron concentration, see i.e. Druilhet et al. (1971).

Fourier analyses of the time variations of radon and thoron

Periodic and high-dependent theoretical solutions are derived for the disturbances of the concentrations of radon and thoron in air above the ground surface by Staley (1966). In these solutions sinusoidal oscillations are assumed.

In the present investigation we have studied the diurnal wave of radon and thoron by Fourier analysis. Other periods than the diurnal wave have also been taken into consideration.

In order to obtain spectral estimates of the variations of radon and thoron over a wide range of frequencies lengthy time series of frequent readings are analyzed. A time series $R = f(t)$ of the period T, defined and bounded in the range of $0 \leqslant t \leqslant T$, may be expressed as a Fourier series.

In the present investigation R represents a recorded radon or thoron curve. An integration of $R = f(t)$ over the range $0 \leqslant t \leqslant T$ gives the Fourier coefficients

$$a_k = 2/T \int_0^T f(t) \cos\,(2\pi kt/T)\,dt$$

and

$$b_k = 2/T \int_0^T f(t) \sin\,(2\pi kt/T)\,dt$$

where k refers to the kth harmonic.

The amplitude of the kth harmonic, viz. $R_k = \sqrt{(a_k^2 + b_k^2)}$, is now calculated for different k-values. In order to obtain a more general expression of the amplitudes a relative amplitude is determined. The relative amplitude is here expressed a $R_k/\bar{R}$, where $\bar{R}$ is the mean-value of the observation series.

The phase angle v_k is given from the expression $\tan v_k = {}^a k/b_k$.

Results of our Fourier analyses of the variations of radon and thoron in the air at the height 0.3 m can be seen in Tables I*a* and I*b*.

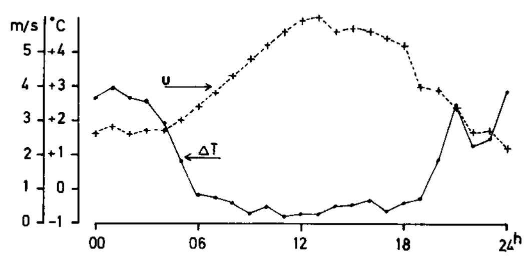

Fig. 9. Diurnal variations of wind speed and temperature stratification.

Table I. *Relative amplitudes* ($R(T)/\bar{R}$) *of the first six harmonics of the diurnal wave of radon and thoron at the height 0.3 m*

T represents the period of the harmonic and $\bar{R}$ is the mean-value of the observation series

Period T in hours	24	12	8	6	4.8	4
Table I*a* *Four days with near-neutral stratification*						
$R(T)/\bar{R}$ in %						
Radon	21	2	7	3	3	2
Thoron	5	6	4	8	6	6
Table I*b*. *Five days with fine-weather*						
$R(T)/\bar{R}$ in %						
Radon	69	26	9	4	4	4
Thoron	34	14	15	11	8	12

Table I*a* represents conditions with near-neutral stratifications and Table I*b* conditions with unstable stratifications, viz. fine weather with unstable stratification in the daytime and stable stratification at night.

Periods shorter than 12 hours have greater relative amplitudes $R(T)/\bar{R}$ for thoron than for radon. For longer periods the relative amplitudes of radon are greter than those of thoron.

The diurnal variation of radon can thus be represented by a few harmonics. For thoron, however, we need several harmonics.

These differences between radon and thoron probably depend on the different disintegration constants of the two nuclides (3.8 days and 54.5 sec respectively).

Great turbulent exchanges in situations with unstable stratification and a small vertical transfer of natural radioactivity under conditions with stable stratification probably cause the great amplitudes of radon and thoron for the diurnal wave.

On overcast days (near-neutral stratification) the relative amplitudes of the variations are small. Especially thoron seems to lack a diurnal wave.

Table II represents measurements of radon and thoron at the heights 0.05 and 3 m under fine weather conditions. For especially thoron the relative amplitudes are greater at the height 0.05 m than at the height 3 m. Like the previous tables the diurnal wave is more pronounced for radon than for thoron.

The amplitude of the diurnal wave of radon and thoron decreases with increasing height.

The phase of the diurnal wave of radon seems to increase somewhat with increasing height. This means that the time of maximum radon concentration becomes later with increasing height. From our measurements the change of phase of the diurnal wave between the heights 0.05 m and 3 m becomes 0.2 hours (standard deviation = 0.2 hours).

For thoron the change of phase of the diurnal wave becomes about −1.4 hours (standard deviation = 1.4 hours). The great standard deviation can be explained by the fact that diurnal variation of thoron is represented by many harmonics; the more the larger heights.

The structures of the time-variations of the α- and β-activity and the radon and thoron concentrations in the atmospheric surface layer will be studied more penetratingly in later papers. Auto-correlation functions of the natural α-activity are presented in a previous paper, Israelsson et al. (1972*a*).

Summary

In order to achieve continuous measurements of radon and thoron at the ground level measurements were made of the radon and the thoron contributions to the ionization in an ionization chamber by a method similar to those given by Norinder et al. (1952) and Israel (1964). With a special electronic equipment the concentrations of radon and thoron can be measured every 15 min.

Our measurements indicated that the intake tube can disturbe the natural conditions, i.e. incorrect profiles may be obtained quite near

Table II. *Relative amplitudes* ($R(T)/\bar{R}$) *of the first five harmonics of the diurnal wave of radon and thoron at the heights 0.05 m and 3 m under 6 fine weather days*

T represents the period of the harmonic and $\bar{R}$ is the mean-value of the observation series

Period T in hours		24	12	8	6	4.8
$R(T)/\bar{R}$ in %						
Radon	0.05 m	83	22	9	7	6
	3 m	81	23	7	12	5
Thoron	0.05 m	27	12	9	6	7
	3 m	19	15	21	12	11

the ground surface. By using a tube containing several holes these disturbances can practically be eliminated.

Simultaneous measurements of the vertical distributions of radon and thoron are made. Due to the short half-life time (54.5 s) the thoron concentration decreases rapidly with height. Stable stratification at night-time causes remarkable gradients near the ground surface. In daytime with unstable stratification and a considerable vertical transfer the gradients of thoron are much smaller. At heights of about 1 m the differences in the thoron concentration between night and day are fairly small.

Radon, however, with longer half-life time (3.8d) shows smaller vertical gradients than thoron. Stable stratification and inconsiderable upward transfer causes high radon contents at all heights.

From the studies of the variations of radon and thoron concentration with time we observed a pronounced diurnal variation. A minimum in daytime caused by a great turbulent transfer and a great concentration value at night-time with stable stratification and a small vertical exchange in the surface layer. The diurnal variation of thoron is less pronounced than the variation of radon probably due to the different magnitudes of their decay constants.

Fourier series analysis of the variations of radon and thoron shows greater variations of thoron for periods shorter than 12 hours. For longer periods, however, the analysis indicates greater amplitudes for radon. These differences may also be due to the different decay constants of radon and thoron.

In later papers we will treat the structures of the time-variations of the α- and β-activity, radon and thoron concentrations more penetratingly by using numerical filter methods. Furthermore results from simultaneous measurements of eddy exchange coefficients of radon, thoron and momentum will be presented.

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ОДНОВРЕМЕННЫЕ ИЗМЕРЕНИЯ РАДОНА (Rn^{222}) И ТОРОНА (Rn^{220}) В ПРИЗЕМНОМ СЛОЕ АТМОСФЕРЫ

С помощью аппаратуры, аналогичной использовавшейся Нориндером и др. (1952) и Израэлем (1964), были проведены одновременные и прямые измерения содержания радона и торона вблизи поверхности земли. Представлены данные по суточным изменениям и вертикальным профилям концентраций радона и торона. Согласно измерениям вертикальные градиенты радона и торона максимальны вблизи поверхности почвы ночью. Соотношения между концентрациями радона и торона изменяются в среднем от 0,1 до 3,0 в слое до высоты 3 *м*. Произведен анализ Фурье временных изменений этих концентраций. Суточная волна для радона представляется немногими гармониками. В случае торона, однако, необходимо большее количество гармоник.