Diurnal variations of radon and thoron decay product concentrations in the surface layer of the atmosphere and their washout by precipitations

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ABSTRACT

Data on diurnal variations of radon and thoron decay product concentrations in the surface layer of the atmosphere in the Moscow region are given. It is noted that the maximum of radon exhalation from the soil occurred at night. (Measurements were carried out in summer in dry weather.) The strongest upset of radioactive equilibrium between radon daughter products was observed in the first half of night and in the day-time.

From the degree of non-equilibrium between RaA:RaB:RaC during the rain the constant of their washout from the lower layer of the troposphere was estimated. The effect of radon and thoron exhalation variations on variations of their concentrations in the surface layer of the atmosphere is theoretically considered.

At the present time there are many works devoted to researches of diurnal variations of radon and thoron decay product concentration in the surface layer of the atmosphere (STYRO, 1959; BLIFFORD *et al.*, 1956; SERVANT, 1965). In most papers the form of diurnal course of radon and thoron decay product concentrations and its dependence on meteorological conditions is considered. But some not less interesting and important aspects of the problem were either not studied at all or studied very little. In our paper we touch some of them. These are first of all, research of diurnal variations of radioactive balance between short-lived radon decay products in the surface layer of the air and the estimation of the effect of exhalation variations on changes of radon and its decay product concentration in the atmosphere. Besides, in the paper the most common variations of the diurnal course of radon and thoron concentrations with the time of year are established. We also made an attempt to estimate the diurnal course of radon exhalation variations.

1. Diurnal variations of radon exhalation

The value of radon exhalation from the soil was determined by the following method. Rn was accumulated in the cylindrical chamber placed on the soil surface by the open end. This method was described in detail in the literature (STYRBO, 1959; SSISSIGUINA, 1965). The time of accumulation is one hour. As a rule 4 or 5 chambers were used simultaneously. They were placed on the soil at a distance of 2–4 m from one another. The results of measurements were averaged. Radon after being accumulated was transferred into the chamber of standard scintillation α-radiometer, its brand is PAA-1. This radiometer allowed to measure directly in the chamber the radon concentration of the order 10^{-12} curie and higher. Calibration of the device was carried out with the help of a standard source having $5 \cdot 10^{-10}$ gram of radium. Measurements were conducted 3 hours after the introduction of radon into the radiometer chamber, that is, after establishing of radioactive balance between radon and its decay products. The computation of the exhalation value E was made according to the formula:

$$E = \Delta Q/St, \quad S = V/h,$$

where h is the height of the air layer in which radon exhalated from the soil was accumulated (in our measurements $h = 3$ cm; it is a distance

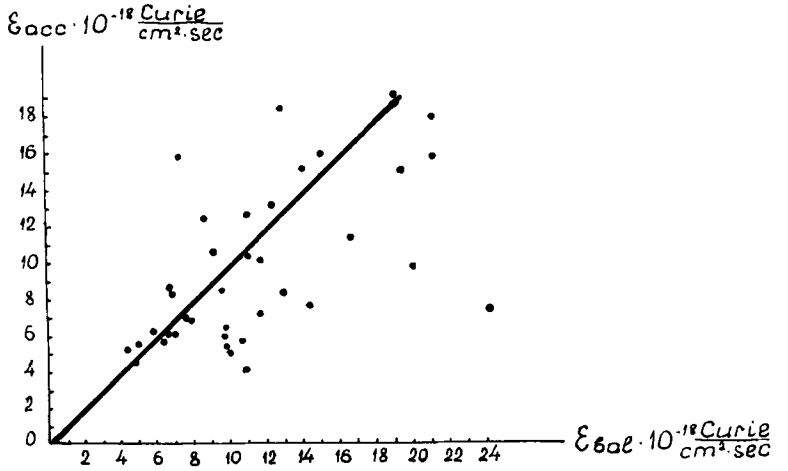

FIG. 1. Comparison of exhalation values determined by accumulation and balance methods (E_{acc}; E_{bal}).

from the soil surface to the partition in the accumulation chamber), v is the volume of the examined sample, t is the time of accumulation, Q the increase of radon amount at the accumulation chamber during the time t: $Q = Q_t - Q_0$. Q_0 which is proportional to the radon concentration at the soil surface at the moment of placing chamber does not usually exceed 10–20% of Q_t. The statistical error in determination of radon concentrations in the radiometer chamber was not higher than 10%.

The method of accumulation has some significant drawbacks. The main of them is that this method isolates the studied part of the soil from the atmospheric air movements. Therefore in a number of cases this method was used simultaneously with the method of balance for calculating the value E.

For friable rocks with homogeneous distribution of radium with uniform density and with established radon flow from the soil into the atmosphere the equation of balance in the column cross section of which is equal to the unit area can be written:

$$\lambda_0 Q = E + Q_i \lambda_0 \quad \text{or} \quad E = (Q - Q_i)\lambda_0.$$

Here $Q = q\infty$. H, supposed total amount of radon in the soil in the column without exhalation the height of which is H; $q\infty -$, radon concentration at a depth of 2 m where it is considered established and nonchanged with depth; Q_i, actual amount of radon in the studied column of the soil. For Q_i and Q determination the measurements of radon concentrations in the soil at a depth of 10, 25, 37, 50, 75, 100 and 200 cm were carried out. Air was sampled at a given depth with the help of special sounding duraluminium tubes 8 mm in diameter with drilled holes at the end. Drilling was performed with augers 25 mm in diameter. The curve of vertical distribution of radon concentrations in the soil was divided into the parts with linear change of concentration and the value Q_i was determined as follows:

$$Q_i = \sum_{x=1}^{\Omega} \frac{q_x + q_x + 1}{2} h_i,$$

where q_x and q_{x+1} are values of concentrations at lower and upper boundary of the layer the width of which is h_i.

Results of comparison of E measurements by these two methods are shown in Fig. 1. There were all in all 8 series of twenty-four-hour measurements of radon exhalation from the soil in the summer. Fig. 2 shows measurements averaged from three series of observations during the period from the sixth through the sixteenth of August in 1963. Measurements were carried out in sand soils in the Moscow region in dry weather under anticyclone conditions. In most series there was observed a natural change of radon flow out of the soil with time, with a maximum at night hours (24–04 h) and a minimum ratio of the magnitude of 1.5–2.0.

Temperature gradients in the soil at depths of 10 and 40 cm are given for comparison in Fig. 2.

In the period of maximum E values at night hours minimum radon concentrations in the soil are observed in the layer down to 25 cm. But in the layer lower than 40 cm it is quite the contrary: maximum radon concentrations in the soil occur at night hours.

Periodicity of exhalation value variations is due to diurnal variability of some factors responsible for the process of interchange of gases between the soil and the atmosphere. Among these factors the most important are temperature of the upper layer of the soil (approximately down to 40 cm) and the intensity of vertical turbulent exchange in the surface layer of the air. Increase of turbulent mixing intensity promotes the radon outflow from the soil-atmosphere interface and the increase of exhalation. On the other hand, cooling of the higher layer of the soil at night hours while at lower layers temperature remains higher (Fig. 2) can promote exchanging the soil air along the vertical and bringing the radon to the soil surface. Due to this radon flow from the soil into the atmosphere is increased at night hours. Higher levels of exhalation at the day time hours in comparison to the evening ones, on the contrary, are connected with the influence of turbulent exchange in the surface layer of the atmosphere on the magnitude of exhalation. Minimum values of E in the evening hours, on the one hand, correspond to relatively low intensities of turbulent exchange and, on the other hand, to the absence of temperature gradients in the soil with depth.

The attempt to establish connections between the exhalation value and change of the atmospheric pressure was not successful. And on the contrary the influence of precipitations is clearly seen. After rain, as a rule, exhalation value and gradients of radon concentration in the soil with depth were sharply diminished.

2. Diurnal variations of radon and thoron decay product concentrations

Measurements of radon and short-lived radon and thoron decay product concentrations in the surface layer of the atmosphere were carried out in the Moscow region in the summer

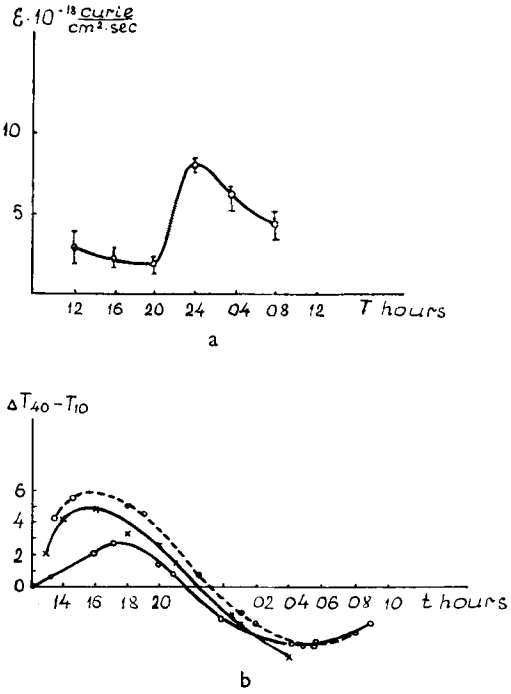

FIG. 2. *Top:* diurnal variations of radon exhalation averaged over 3 days (72 hours). *Bottom:* the diurnal course of the temperature gradient in the soil during these three days (between 10 and 40 cm).

(June–August) of 1963 and 1964. To determine radon concentrations the method of its adsorption on activated carbon was used (STYRBO, 1959; SSISSIGUINA, 1964). To measure radon decay product concentrations (from α-radiation) the method of air filtration through fine filter was used. The time of filtration was 20 minutes. Concentrations were calculated according to formulas of accumulation and decay of radon daughter products on filter (BARANOV & GORBUSHINA, 1962), assuming the equilibrium between them in the atmospheric air. The time of air filtration when determining ThB + ThC concentrations was 2 hours. ThB + ThC concentration calculation was made analogous to that in the paper by MALAKHOV & CHERNYSHOVA. The height of sampling is one and eleven meters above the ground.

Results of measurements of radon and thoron decay product concentrations averaged for 2 hour intervals are presented in the Table 1. It is noted the well-known diurnal course which has the form of a simple wave with maximum in the morning hours (4–6 hr) and minimum

TABLE 1. *Mean-hour concentrations of radon and thoron decay products (10^{-15} curie/liter)in the surface layer of the atmosphere. (Moscow region, Summer, 1964.)*

hr ... Isotopes	0–2	2–4	4–6	6–8	8–10	10–12	12–14	14–16	16–18	18–20	20–22	22–24
RaB + RaC	160	200	258	250	187	120	122	118	123	144	155	158
ThB + ThC	3.8	4.8	5.7	5.5	3.5	1.9	2.0	1.7	2.5	2.9	3.5	3.6
$\frac{\text{RaB + RaC}}{\text{ThB + ThC}}$	42	41.5	44.5	45.6	53.5	63.3	61.1	69	49.6	49.7	44.5	44

in the afternoon hours (14–16 hr). Somewhat unexpected was the diurnal course of the concentration ratio RaA + RaC/ThB + ThC having maximum values in the daytime (14–16 hr) and minimum ones at night (0–2 hr). For stationary state, quite reasonably supposing that it is ThB diffuses from the soil and not thoron, the ratio Rn/ThB can be expressed by the formula:

$$\frac{\text{Rn}}{\text{ThB}} = \frac{E_{\text{Rn}}}{E_{\text{ThB}}} \frac{\sqrt{\lambda_{\text{ThB}}}}{\sqrt{\lambda_{\text{Rn}}}} \exp\left[-\frac{1}{K}(\sqrt{\lambda_{\text{Rn}}} - \sqrt{\lambda_{\text{ThB}}})\right].$$

Here λ_{ThB} and λ_{Rn} are decay constants, K is the coefficient of turbulence constant with height, E_{Rn} and E_{ThB} values of exhalation. Since $\lambda_{\text{Rn}} < \lambda_{\text{ThB}}$, the ratio Rn/ThB must increase with decrease of K. But quite the opposite picture is observed. It is due to the fact that diurnal variations of radon and thoron exhalation are somewhat different, or that the diffusion processes are nonstationary, or that radon decay products at different times of the day deviate in various degrees from the radioactive balance with radon.

To clear up the role of the seasonal factor in changes of diurnal variations of radon and thoron decay product concentration we treated the data of measurements in Vienna in 1964 which were received by the Moscow World Center of data on the program of International Geophysical Cooperation. In Vienna they used the method of filtration of air. The duration of filtration was 3 hours. Measurements of filter β-activity were carried out in 5 minutes, 2.5 hours and 5 days. Values of RaB + RaC and ThB + ThC concentrations in the air were calculated by us analogous to those made in the paper by MALAKHOV & CHERNYSHOVA (1965). And it was considered that the contribution of β-radiation of fission products into the results of the first and second measurements was equal to the intensity of β-radiation of the sample in 5 days. When making an analysis we considered

TABLE 2. *Change of diurnal variations of radon and thoron decay product concentration with time of year (measurements in Vienna).*

Month, 1964	RaB + RaC		RaB + RaC	ThB + ThC		ThB + ThC
	Time of max	Time of min	$\frac{\text{max}}{\text{min}}$	Time of max	Time of min	$\frac{\text{max}}{\text{min}}$
I	5.8	11–14	1.4	2.5	11–14	1.5
II	1–1	14–17	1.3	5–8	14–17	1.4
III	2–5	14–17	1.2	2–5	14–17	1.2
IV	5.8	14–17	2.3	2–5	14–17	2.6
V	5.8	14–17	2.9	2–5	14–17	2.5
VI	5.8	14–17	2.0	2–5	14–17	2.5
VII	2.8	14–17	2.7	2–5	14–17	3.0
VIII	5.8	14–17	2.3	2–5	14–17	3.1
IX	2.5	14–17	2.3	2–5	14–17	3.1
X	5.8	11–14	1.7	23–02	11–14	2.2
XI	5.8	14–17	1.4	2–5	11–14	1.4
XII	—	—	1.0	23–5	11–14	1.1

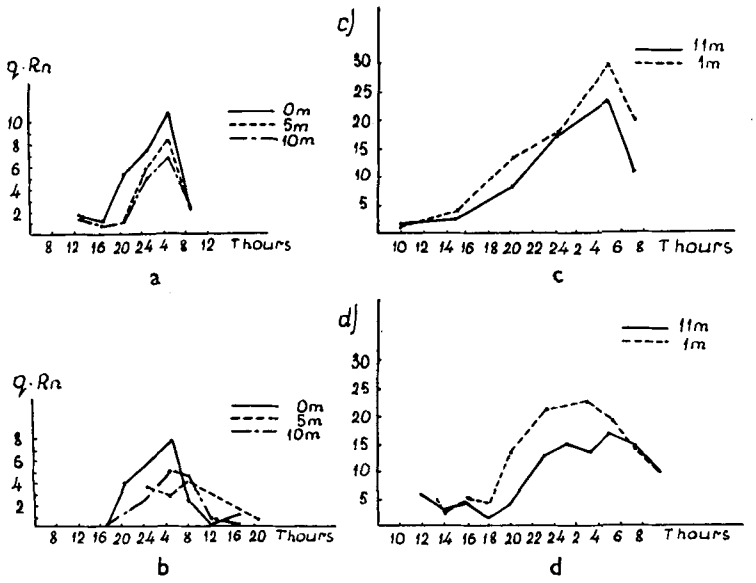

FIG. 3. Diurnal variations of Rn (*a*, *b*) and ThB (*c*, *d*) concentration at three various heights in the surface layer of the atmosphere.

the diurnal course obtained by averaging of all the data of measurements carried out during every month. Results given in Table 2 in general agree with the data of our measurements (Table 1). The amplitude of diurnal variations (ratio of maximum concentrations to minimum ones) increases almost 3 times during the period from the winter to the summer season. On the other hand, the amplitude of diurnal variations is approximately the same for radon decay products and thoron decay products which indicates that there is community of causes of their diurnal variation course. The diurnal change of the ratio RaB + RaC/ ThB + ThC in the summer occurs analogous to the data from Table 1. In the winter diurnal variations of this ratio are almost smoothed.

During 7 series of 24-hour measurements in the Moscow region in the summer of 1964 the data of diurnal course of radon and thoron concentrations on 3 levels above the ground, 0 m, 5 m and 10 m, were obtained (Fig. 3). In all the cases the most significant differences in concentrations with height were observed at night, the amplitude of diurnal variations decreasing with the increase of height above the ground. In three cases out of seven there was noted some delay in appearance of concentration maximum at higher levels but this delay was not more than 3 hours for the denoted altitudes.

STENHEY, MOSES & DUCAS (1960) give the data of 3 day (72 hour) measurements of radon concentrations at 4 altitudes from 0.32 cm to 39.9 m. Results of these measurements are presented in the Table 3. They also show that the amplitude of radon concentration diurnal course gradually decreases with height. Delay

TABLE 3. *Diurnal variations of radon concentration in the layer from 0 to 40 m.*

Date ...	20–21. IX. 1958					16–17. VII. 1958				27–28. VII. 1958		
Altitude (m) ...	0.97	5.7	23.8	39.9	0.32	0.97	5.7	39.9	0.32	0.97	23.8	39.9
Time of maximum	3	3	2	2	24	24	3	6	23	1	6	6
Time of minimum	10	11	10	10	10	12	10	10	16	16	16	16

in appearance of maximum at a level of 39.9 m relative to that of 0.97 m in two cases out of three reached 5–6 hours.

The time of establishing minimum concentrations was the same for all the heights with rare exceptions.

3. Radioactive equilibrium between radon short-lived decay products

Separate determination of RaA, RaB and RaC concentrations in the surface air was carried out by analysing the curve of radioactive decay of aerosol samples collected on the filter FPP-15 (from α-radiation).

Decay of mixture consisting of radon short-lived decay products can be described by the following equation:

$$Ae^{-\lambda_1 t} + Be^{-\lambda_2 t} + Ce^{-\lambda_3 t} = D_t.$$

Here λ_1, λ_2, λ_3 are the decay constants for RaA, RaB, RaC; A, B, C are constants depending on the amount of RaA, RaB, RaC on the filter at the moment $t = 0$ (ending of filtration). D_t is the absolute velocity of count at the moment t. If to multiply the right part and the left one of the equation by $e^{\lambda_2 t}$, it is possible to obtain

$$Ae^{(\lambda_2-\lambda_1)t} + B + Ce^{(\lambda_2-\lambda_3)t} = De^{\lambda_2 t}.$$

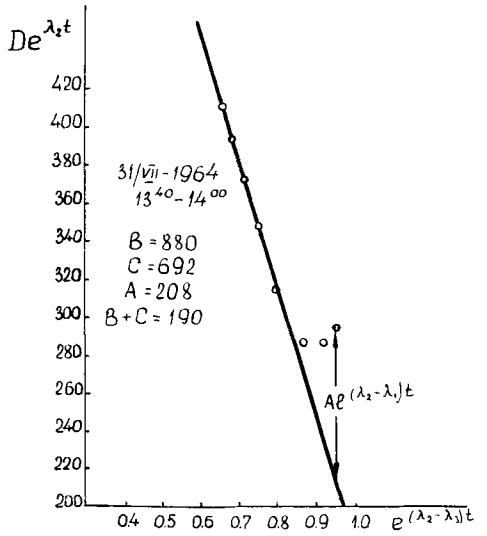

FIG. 4. A typical case of analysis of RaA + RaB + RaC mixture decay curve.

The term $Ae^{(\lambda_2-\lambda_1)t}$ decreases relatively fast with increase of t, and at $t > 10$–15 min it is practically equal to nil. Then the remaining part of the equation will be the equation of a straight line for the variables $e^{(\lambda_2-\lambda_3)t}$ and $De^{\lambda_2 t}$ (MALAKHOV, 1964). At the points where $e^{(\lambda_2-\lambda_3)t} = 1.0$ or $e^{(\lambda_2-\lambda_3)t} = 0.0$ ordinates of this straight line are equal to $B + C$ and B, respectively.

Hence it is possible to determine B and C. The difference between $De^{\lambda_2 t}$ and the corresponding ordinate of direct line at any $t < 10$–15 min gives the value of $Ae^{(\lambda_2-\lambda_1)t}$ from which it is easy to determine A (Fig. 4).

Using the well-known formulas for accumulation of RaA, RaB and RaC on the filter at the process of filtration and formulas of these isotopes decay after the end of filtration (BARANOV & GORBUSHINA, 1962), it is possible to express constants A, B, C through the concentrations of RaA, RaB, RaC (q_1, q_2, q_3) in the surface layer of the air.

$$q_1 = 0.9770\ A/[1 - \exp(-\lambda_1\theta)]\ V;$$

$$q_2 = 0.2642\ B/[1 - \exp(-\lambda_2\theta)]\ V - q_1 \cdot 1.1287;$$

$$q_3 = C/[1 - \exp(-\lambda_3\theta)]\ V + 2.785\ q_2 + 3.3956\ q_1.$$

Here V = velocity of filtration in volume units for a minute; θ = time of filtration. These formulas are obtained for case when α = activity of filters (RaA + RaC) is measured and when it is assumed that the decay of atom of RaC′ follows practically instantaneously the decay of RaC atom. And besides registration efficiency of α-radiation from RaA and RaC′ by the counting device considered equal. From the given formulas and knowing A, B and C it can be found q_1, q_2, q_3. To receive more exact values the value of A was usually determined by making additional curves. For this in semilogarithmic scale (abscissa is time in minutes, ordinate is $\lg[Ae^{(\lambda_2-\lambda_1)t}]$) the points were plotted corresponding to the difference between experimentally measured values of $De^{\lambda_2 t}$ and the direct line at $t < 10$–15 min. Then through these points a straight line was drawn with the slope corresponding to the RaA half-life. Extrapolation of the received straight line to $t = 0$ allowed to determine A.

In our paper the ratio of concentrations of RaA:RaB:RaC was determined, RaA concentration being assumed equal to 1. We used

TABLE 4. *Radioactive balance between short-lived radon decay products.*

hr ... Isotopes	0–2	2–4	4–6	6–8	8–10	10–12	12–14	14–16	16–18	18–20	20–22	22–24
RaA	1	1	1	1	1	1	1	1	1	1	1	1
RaB	0.5 ±0.07	0.9 ±0.06	0.8 ±0.07	0.9 ±0.05	0.8 ±0.05	0.7 ±0.07	0.4 ±0.08	0.7 ±0.07	0.6 ±0.07	0.6 ±0.05	0.5 ±0.08	0.4 ±0.08
RaC	0.4 ±0.08	0.7 ±0.07	0.7 ±0.08	0.9 ±0.07	0.7 ±0.07	0.7 ±0.06	0.4 ±0.08	0.6 ±0.07	0.5 ±0.06	0.5 ±0.09	0.4 ±0.08	0.3 ±0.08
Number of measurements	(22)	(22)	(16)	(21)	(20)	(24)	(33)	(18)	(17)	(20)	(25)	(21)

260 measurements carried out in 1963 and 1964 during the period from June through August. Measurement data were divided for two hour intervals and were averaged. The results are presented in Table 4. Mean-root-square deviations are also shown. The amount of the measurements is given.

The most strong disequilibrium between radon decay products takes place in the evening and during the first half of night. The sharp upset of radioactive equilibrium occurs also immediately after noon. Radon decay products approach most closely to the equilibrium in the morning.

During the analysis of separate cases it was found out that sharp upset of radioactive equilibrium between RaA, RaB and RaC was established during change of weather from windy one to complete calm. Thus on the 14th of August in 1964 from 16.40 to 17.00 the ratio of radon decay product concentrations was 1:0.9:0.8. Directly after the establishing of calm from 19.40 to 20.00 this ratio changed; it became 1:0.05:0.02. And then it increased and reached the value 1:0.7:0.7 by 22.00. Establishing of calm was accompanied by increase of radon decay product concentrations. To all appearances, sharp decrease of intensity of vertical turbulent exchange promotes the accumulation of radon in the surface layer and the corresponding upset of radioactive equilibrium. Hence, the disequilibrium in the evening and in the first half of night, that is during the period when intensity of vertical turbulent exchange usually sharply decreases, is quite understandable. It is very difficult so far to explain whether the disequilibrium at noon (12–14 hr) is connected with other causes. It is most likely due to frequent appearance and destruction of convective vertical flows at this time of the day (SERVANT, 1965; KIRICHENKO, 1962; SHOPAUSKAS, 1965).

In one of the cases sharp upset of equilibrium between RaA, RaB, RaC was observed in the morning after night frosts. If early in the morning the ratio of RaA, RaB, RaC concentrations was 1:1:1, by 10 o'clock in the morning when the ground was thawed out and dried this ratio dropped to 1:0.2:0.1. It is apparently explained by the appearance of new portions of radon without its decay products from the soil the capillaries of which became free of ice and water.

The effect of precipitation on the extent of radioactive disequilibrium between radon short-lived decay products was specially studied. It was established that in many cases this effect was significant. In the morning hours in the winter and spring of 1963–1964 two series of measurements of RaA, RaB, RaC concentration ratio in the surface air during precipitation and in dry weather were carried out. From 27 measurements in dry weather radioactive equilibrium (not lower than 1:0.9:0.9) was observed in 22 cases. RaA, RaB, RaC mean concentration ratio was $1:0.9 \pm 0.03:0.9 \pm 0.06$. (Mean-root-square deviations from arithmetical mean are presented.)

From 15 measurements during precipitation the ratio close to radioactive equilibrium was only in 5 cases. Using the obtained data it is possible to attempt to estimate separately the constant of washout of RaB and RaC. It is not difficult to show that for established state the RaA, RaB, RaC concentration ratio (in activity units) in case of precipitations can be expressed according to the laws of kinetics of first order as follows:

$$q_1 : q_2 : q_3 = 1 : \frac{\lambda_2}{\lambda_2 + G_2} : \frac{\lambda_2 \lambda_3}{(\lambda_2 + G_2)(\lambda_3 + G_3)}.$$

Here λ_2 and λ_3 are decay constants of RaB and RaC; G_2 and G_3 are washout constants of RaB and RaC. Use of the above formula gives approximately equal values of $G_2 = (1.5 \pm 0.5) \cdot 10^{-4}$ sec^{-1} and $G_3 = (1.0 \pm 0.51) \cdot 10^{-4}$ sec^{-1}, though the washout constant for RaB is a little more than for RaC. G_2 and G_3 characterize washout of radon decay products by falling precipitation particles. Their values are several times lower than those of washout constant of RaA and RaC from the cloud air by cloud drops, estimated by STYRO B. I. with coworkers (1964). On the other hand, the received values of G_2 and G_3 are two to four times higher than those of washout constant of RaB + RaC, determined by one of the authors from measurements on the slope of Elbrus (MALAKHOV & SOLODIKHINA, 1962) and need to be thoroughly examined.

For estimation of ThB washout from the surface layer of the atmosphere by falling precipitation particles the case of long-term rain was used when for 3 hours ThB concentration in the surface air decreased to 0.6 of its level before the beginning of precipitation. If we consider that the decrease of concentration during precipitation occurred according to the exponential law, the value $1.4 \cdot 10^{-4}$ sec^{-1} will be obtained for ThB washout constant. In all the other cases the duration of precipitation was considerably less than the duration of collecting the sample and therefore there was no sense in determination of the washout constant.

It is interesting to note that in a number of papers (SERVANT, 1965; FONTAN & BIROT, 1964) the authors do not find during precipitation the significant decrease of the concentration of all the radon short-lived decay products in comparison to the radon concentration. It can be explained by the fact that the total concentration of radon decay products especially determined from α-radiation is changed less under the influence of precipitation than the ratio of separate radon short-lived decay product concentrations.

4. Variations of natural radioactivity in the atmosphere caused by change of Rn and Th exhalation

Periodical change of radon and thoron and their daughter concentrations in the surface layer of the atmosphere can be the result of both diurnal variations of intensity of vertical turbulent exchange and diurnal variations of exhalation. We shall consider below the effect of exhalation variations on change of radon, thoron and their daughter concentrations in the atmospheric air. Let us touch, first of all, the periodical variations of exhalation. The equation of radon and thoron diffusion for nonstationary conditions can be written in the following form:

$$\frac{\partial q_0}{\partial t} = \varkappa \frac{\partial^2 q}{\partial z^2} - \lambda_0 q_0. \tag{1}$$

Here q_0 = radon (or thoron) concentration, λ_0 = the constant of its decay, z = height, t = time, $\varkappa$ = coefficient of vertical turbulent diffusion. The latter is constant with height and during the day. The last condition allows us to estimate better concentration change under the influence of exhalation change. We solve the equation for boundary conditions:

$$q_0 \to 0 \text{ at } z \to \infty, \quad \varkappa \left.\frac{\partial q_0}{\partial z}\right|_{z=0} = E(1 + \varepsilon \cos \omega t),$$

where $E(1 + \varepsilon \cos \omega t)$ = diurnal exhalation variations presented as a sum of exhalation constant level and cosinusoidal variations. We are interested in the stationary state when it is possible to neglect the effect of initial conditions. Then the solution can be presented as a sum of two terms: $q_0 = q_0^{\text{stationary}} + q_0^{\text{variable}}$. Here q_0^{stat} is a solution of stationary ($\partial q/\partial t = 0$) equation of radon diffusion corresponding to constant level of exhalation, q_0^{var} describes established variation of radon (or thoron) concentrations under cosinusoidal variations of exhalation. The boundary condition for the second term of the solution can be changed by

$$\varkappa \left.\frac{\partial q_0}{\partial z}\right|_{z=0} = E\varepsilon e^{i\omega t}$$

and it is necessery to find solution for q_0^{var} in the form of $Re(Ae^{\alpha z + \beta t})$. Then we obtain

$$q_0^{\text{var}} = \frac{\varepsilon E}{\sqrt{\varkappa}\sqrt[4]{\lambda_0^2 + \omega^2}} \exp\left(-\sqrt{\frac{\sqrt{\lambda_0^2 + \omega^2}}{\varkappa^2}}\, z \cos\frac{\varphi_1}{2}\right)$$

$$\times \cos\left[\omega t - \sqrt{\frac{\sqrt{\lambda_0^2 + \omega^2}}{\varkappa^2}}\, z \sin\frac{\varphi_0}{2} - \frac{\varphi_0}{2}\right), \tag{2}$$

where $\varphi_0 = \text{arc tg } \omega/\lambda_0$, q_0 in atoms per unit volume.

The multiplier

$$\exp\left(-\sqrt[4]{\frac{\lambda_0^2+\omega^2}{\varkappa^2}}\, z \cos\frac{\varphi_0}{2}\right)$$

shows the decrease of diurnal variation amplitude and cosine delay of variations with the height above the ground. The value of q_0^{stat} is known:

$$q_0^{\text{stat}} = \frac{E}{\sqrt{\lambda_0 \varkappa}} \exp\left(-z\sqrt{\lambda_0/\varkappa}\right).$$

For radon (thoron) decay products differential equations were solved

$$\frac{\partial q_n}{\partial t} = \varkappa \frac{\partial^2 q_n}{\partial z^2} - \lambda_n q_n + \lambda_{n-1} q_{n-1} \qquad (3)$$

for the boundary condition

$$\varkappa \left.\frac{\partial q_n}{\partial z}\right|_{z=0} = 0, \quad n \geqslant 1,$$

that is, it was assumed that only radon (or thoron) without its daughters exhales from the soil and "dry" deposition of radon (thoron) decay products is neglected. Then

$$q_n = q_n^{\text{stat}} + E\frac{\prod_{\varkappa=0}^{n-1}\lambda_\varkappa}{\sqrt{\varkappa}} \times \sum_{i=0}^{n} \frac{1}{\sqrt[4]{\lambda_i^2+\omega^2}\prod_{\varkappa=0\,\varkappa\neq i}^{n}(\lambda_\varkappa-\lambda_i)} \times \cos\left[\omega t - \sqrt[4]{\frac{\lambda_i^2+\omega^2}{\varkappa^2}}\, z \sin\frac{\varphi_i}{2} - \frac{\varphi_i}{2}\right]. \qquad (4)$$

Here $\varphi_i = \text{arc tg } \omega/\lambda_i$;
q_n is in atoms per unit volume.

$$q_n^{\text{stat}} = \frac{E\prod_{\varkappa=0}^{n-1}\lambda_\varkappa}{\sqrt{\varkappa}} \sum_{i=0}^{n} \frac{\exp\left(-z\sqrt{\lambda_i/\varkappa}\right)}{\sqrt{\lambda_i}\sum_{\varkappa=0\,\varkappa\neq i}^{n}(\lambda_\varkappa-\lambda_i)}$$

For studying the effect of separate sharp exhalation variations of radon or thoron on the magnitude of their concentration in the surface layer of the air, equation (3) was solved under the following boundary and initial conditions:

1) $q_{0t} = 0$ at $t = 0$; $\quad \varkappa \left.\frac{\partial q_0}{\partial z}\right|_{z=0} = E$ for $t > 0$;

$q_{0t} \to 0$ at $z \to \infty$.

2) $q_{nt} = 0$ at $t = 0$; $\quad \varkappa \left.\frac{\partial q_n}{\partial t}\right|_{z=0} = 0$;

$q_{nt} \to 0$ at $z \to \infty$, $\quad n > 0$.

The solutions are:

$$q_{nt} = \frac{E\prod_{\varkappa=0}^{n-1}\lambda_\varkappa}{\sqrt{\varkappa}} \sum_{i=0}^{n} \frac{1 - F(z,t,\lambda_i,\varkappa)}{2\sqrt{\lambda_i}\prod_{\varkappa=0;\,\varkappa\neq i}^{n}(\lambda_\varkappa-\lambda_i)}. \qquad (5)$$

Here

$$F(z,t,\lambda_i,\varkappa) = \exp\left(-z\sqrt{\lambda_i/\varkappa}\right)\left[1+\phi\left(\sqrt{\lambda_i t} - \frac{z}{2\sqrt{\varkappa t}}\right)\right] - \exp\left(z\sqrt{\lambda_i/\varkappa}\right)\left[1-\phi\left(\sqrt{\lambda_i t} + \frac{z}{2\sqrt{\varkappa t}}\right)\right];$$

$$\phi(x) = \frac{2}{\sqrt{\pi}}\int_0^x e^{-y^2} dy.$$

If radon (thoron) exhalation equal to E continued during the time θ, then for $t > \theta$

$$q_0 t = \frac{e^{-\lambda(t-\theta)}}{2\sqrt{\pi}} \int_0^\infty \frac{1}{\sqrt{\varkappa(t-\theta)}} \left\{\exp - \frac{(z-\varepsilon)^2}{4\varkappa(t-\theta)} + \exp\left[-\frac{(z+\varepsilon)^2}{4\varkappa(t-\theta)}\right]\right\} \times \varphi(\xi)\, d\xi,$$

$$\varphi(\xi) = \frac{E}{\sqrt{\lambda\varkappa}} F(\xi, \theta, \lambda_0, \varkappa).$$

Now we shall touch some results of calculation made according to the above formulas.

The curves of dependence of radon and thoron concentration increase for various $z_{\text{eff}} = z/\sqrt{\varkappa}$ on exhalation which began at $t = 0$ and was equal to E (formula 5) are shown in Fig. 5. Radon and ThB concentration in the stationary conditions ($t \to \infty$) for corresponding values of z_{eff} was taken for a unit. Very great duration of establishing stationary state attracts attention. For radon, even near the ground surface, stationary state is established 7–10 days after

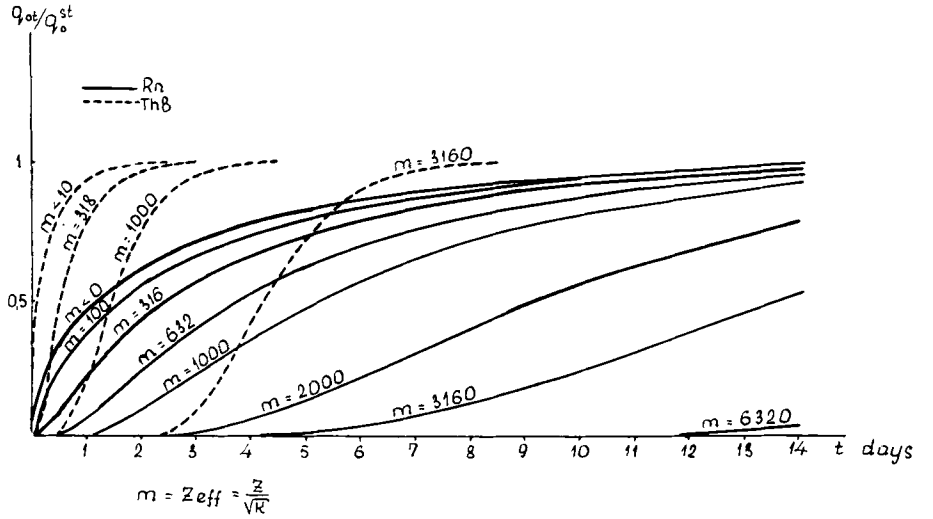

FIG. 5. Increase of radon and ThB concentrations in the atmosphere at $t > 0$ if $E = 0$ at $t < 0$ and if $E = \text{const}$ at $t \geqslant 0$.

the exhalation began. Radon begins to appear at the height $z_{eff} = 623$ sec$^{\frac{1}{2}}$ only 12 hours after its exhalation out of the ground and at the height $z_{eff} = 10^3$ sec$^{\frac{1}{2}}$ in 24 hours. For small altitudes about half the atoms of radon went out of the ground surface during not more than 24 hours and for $z_{eff} = 623$ sec$^{\frac{1}{2}}$ the amount of exhaled atoms reached approximately 7%.

For thoron decay chain (ThB) the stationary concentration establishes about 5–7 times quicker. As in our solution, diffusion is considered on half-line (from $z = 0$ to $z = \infty$) and $\varkappa = \text{const.}$ with height the obtained estimations of time of stationary state establishment should be regarded as estimations of the upper limit. But as for qualitative conclusion of inertial state of radon and thoron concentrations relative to increase of exhalation it is, to all appearances, correct.

Diurnal course of radon concentration in the air caused by diurnal variations of radon exhalation proves to be very small (formulas 2, 4). The amplitude of variations for the extreme case, when $\varepsilon = 1$ and $z = 0$, is only 0.18 from the mean value of concentrations during 24 hours. It decreases with height according to the law $0.18 \exp [0.0073 (z/\sqrt{\varkappa})]$, where $z/\sqrt{\varkappa}$ is expressed in sec$^{\frac{1}{2}}$. When measuring radon exhalation (§ 1) we obtained $E_{max}/E_{min} = 2$, that is, $\varepsilon = \frac{1}{3}$. Corresponding to this case, amplitude of diurnal variations will be 3 times less. Delay of diurnal variations of radon concentration has an hour increase with increase of z_{eff} for about every 40 sec$^{-\frac{1}{2}}$.

The amplitude of ThB diurnal variations at $z = 0$ and $\varepsilon = 1$ is somewhat higher than that of radon: 0.6 of the mean value. Its decrease with height is described as $0.6 \exp [0.0034 (z/\varkappa)]$. Phase delay from exhalation increases about an hour with increase of z_{eff} for about every 40 sec$^{-\frac{1}{2}}$. For about $\varepsilon = \frac{1}{3}$, the amplitude of diurnal course of ThB concentration will be about 20%, that is, the ratio of maximum and minimum values will be 1.5. It is already close to natural ThB diurnal course. Thus, thoron exhalation variations cannot be neglected when considering causes of diurnal course of ThB concentration.

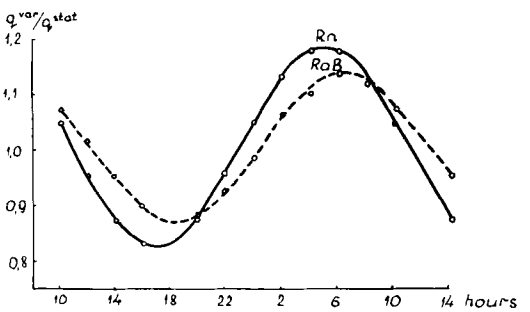

FIG. 6. The diurnal course of radon and RaB concentrations at $z = 0$ calculated theoretically for cosinusoidal exhalation variations and for $K = \text{const}$.

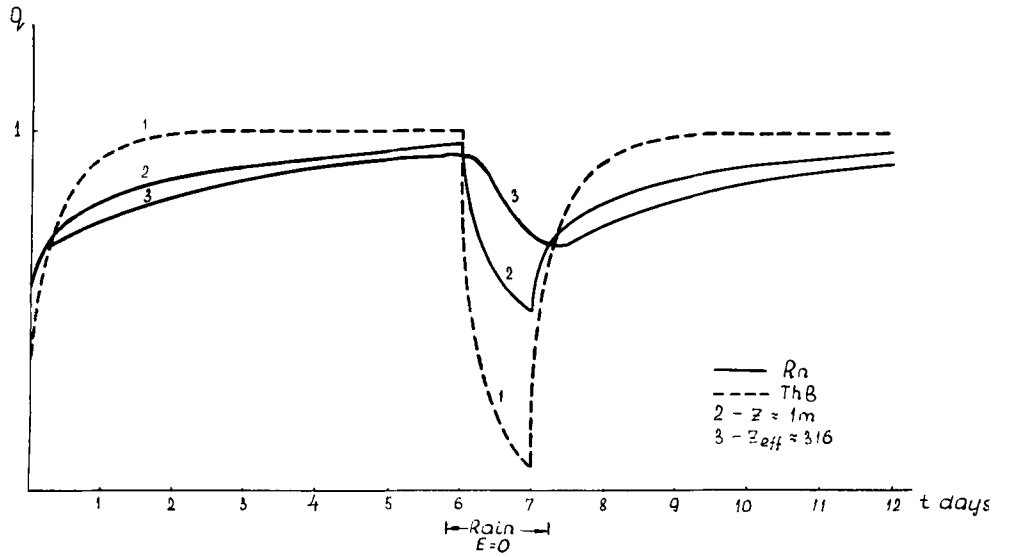

FIG. 7. The established change of radon and ThB concentrations in the atmospheric air provided that every seventh day it is raining ($E = 0$) and on the remaining 6 days exhalation is constant ($E = \text{const}$).

Diurnal course of radon and RaB concentrations ($z = 0$, $\varepsilon = 1$) calculated from the formulas (2) and (4) is presented in Fig. 6 (maximum of E is at 2 o'clock a.m.). RaB variations delay in phase from radon variations. Owing to it, upset of radioactive equilibrium between radon and ThB is the greatest in the first half of night and in the first half of day and minimum differences are in the morning and at the end of the day-time. At night radon concentration is higher than that of RaB and in the day-time the picture is quite the contrary. In general, it agrees qualitatively with our experimental data on concentration ratio of RaA to RaB in the atmospheric air (§ 3). But the observed disturbance of radioactive equilibrium between short-lived radon decay products in the atmosphere is considerably greater than it is expected from calculations according to the formula (3). Besides, on the average in data on concentration ratio of RaA to RaB, given in chapter 2, there are no cases when RaB concentration would be higher than that of RaA. Both these facts, in our opinion, are explained by such a power factor in the atmosphere as periodical variations of intensity of vertical turbulent exchange which are not taken into account in calculations.

Established variations of radon (ThB) concentration are shown in Fig. 7 for the case when every 6 days Rn(Th) exhalation was equal to E and on the seventh day it was nil (it was raining). On the average such relation between dry and rainy times is characteristic of central parts of the USSR (Climatological reference book of the USSR, 1949). Calculation was performed by numeric methods using the formula (5). Delay of radon concentration variation with height attracts attention just as the fact that radon concentration very slowly reaches its level after the finish of precipitations. Maximum radon concentrations near the ground surface constitute 6/7 of the concentration level at constant exhalation under stationary conditions taken for 1.

Calculations according to formula (2) show also that concentration ratio of Rn and ThB has maximum values in the day-time and minimum ones at night, if change of radon and thoron exhalation is synchronous with maximum in the middle of the night and with minimum in the day-time.

Summary

The diurnal course of radon, thoron and their decay product concentrations has maximum early in the morning and minimum in the evening. Its amplitude decreases from summer to winter. The strongest upset of radio-

active equilibrium between radon short-lived decay products is observed in the first half of night and the ratio close to equilibrium is early in the morning (measurements in the summer months). Measurements of radon exhalation showed that its maximum was at night and its minimum was in the evening. The diurnal variations of exhalation led us to the thought to find out their effect on the diurnal course of natural radioactivity of the atmosphere. Theoretical calculations made in the present paper showed that diurnal variations of exhalation can provoke diurnal variations of natural radioactivity parameters which are qualitatively similar to the variations observed experimentally. But quantitatively all these variations are, as a rule, insignificant, except those of ThB. Variations of intensity of vertical turbulent mixing should be acknowledged. Probably the main factor which governs diurnal variations of natural radioactivity in the atmosphere.

Diurnal variations of vertical turbulent exchange are the largest in summer. In accordance with this, the most pronounced diurnal course of radon and ThB concentration occurs in the summer months. The upset of radioactive equilibrium between RaA:RaB and RaC in the first half of night to all appearances is also connected with variations of turbulent exchange. At this time the radon diffusion in the atmosphere along the vertical sharply grows weak, "fresh" radon accumulates in the surface layer. This disturbs the radioactive equilibrium between radon and its decay products especially at the very beginning of the process.

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СУТОЧНЫЕ ВАРИАЦИИ КОНЦЕНТРАЦИИ ПРОДУКТОВ РАСПАДА РАДОНА И ТОРОНА В ПРИЗЕМНОМ СЛОЕ АТМОСФЕРЫ И ИХ ВЫМЫВАНИЕ ОСАДКАМИ

Представлены данные о суточных вариациях концентрации продуктов распада радона и торона и приземном слое в Московской области. Замечено, что максимум выделения радона из почвы приходится на ночь. (Измерения были выполнены летом в сухую погоду.) Наиболее сильное нарушение радиоактивного равновесия между дочерними продуктами радона наблюдалось в первую половину ночи и в дневное время.

По степени неравновесности во время дождя между RaA:RaB:RaC была оценина константа их вымывания из нижнего слоя тропосферы. Теоретически рассматривается влияния изменения выделения радона и торона на вариацию их концентраций в приземном слое атмосферы.