Calculation of planetary distributions of radon and its long-lived daughter concentrations in the troposphere and lower stratosphere

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ABSTRACT

The boundary value problem for the differential equation for mean zonally averaged tracer concentrations in the meridional plane up to 25 km level and from pole to pole is introduced and connected with a model of tracer transport processes in the atmosphere on a global scale. The coefficients of this equation are determined from climatic data on the mean meridional circulation and macroturbulent diffusion. The finite difference analogue of this problem is solved numerically by the method of matrix factorisation, and the annual mean zonally averaged concentration distributions for Rn^{222}, Pb^{210}, and Po^{210} are obtained for several variants of parameters. The results of the calculation are compared with the published data of isotope concentration measurements in the free atmosphere, and the global intensity of macroturbulent diffusion is estimated. The calculated isotope content in the troposphere and stratosphere of the northern and southern hemispheres and their rates of transport across the equator and through the topopause are briefly outlined.

Results of measurements of radon and its long-lived daughter content in the surface air, rain, and free atmosphere lead to several quantitative and qualitative estimates of atmospheric transport and exchange in different scales (Burton & Stewart, 1960; Karol & Vilenskii, 1965; Karol, 1965, 1966; Lambert & Nezami, 1965; Peirson *et al.*, 1966). But the poor quantity of these measurements, especially in the free atmosphere, does not allow an estimate of the planetary pattern of isotope concentrations in the atmosphere.

This paper contains some preliminary results of an attempt to calculate the global distributions of Rn^{222}, Pb^{210}, Bi^{210}, and Po^{210}, average zonal concentrations in the meridional plane from pole to pole and up to 25 km by means of a numerical solution of the boundary problem for the two-dimensional stationary macroturbulent diffusion equation.

As the land surface is the source of atmospheric radon, the planetary distributions of it and its daughter concentrations are three-dimensional. But to obtain the first approximation of the global pattern of these distributions and to estimate the isotope content and exchange in the stratosphere and upper troposphere, it is reasonable to consider the much more simple stationary two-dimensional problem of calculation of zonally and annually averaged concentration distributions.

Constructing a model of radioactive aerosol transport and removal in the atmosphere, and following Davidson *et al.* (1966), Karol (1966), and Reed & German (1965), let us assume that this averaged concentration $c^{(\varkappa)}(r, \theta)$ (atoms in air mass unit) for the isotope $\varkappa$ of the decay chain in the rectangle $(0 < \theta < \pi;\ r_1 < r < H)$ of the meridional plane of spherical coordinate system yields the equation

$$\frac{\partial \prod_r^{(\varkappa)}}{\partial r} + \frac{\partial (\prod_\theta^{(\varkappa)} \sin\theta)}{\sin\theta \partial\theta} + \varrho(\lambda^{(\varkappa)} + \sigma) c^{(\varkappa)} = \varrho \lambda^{(\varkappa-1)} c^{(\varkappa-1)}, \tag{1}$$

where

$$\prod_r^{(\varkappa)} = \varrho(v_r - w) c^{(\varkappa)} - \varrho\left(E_{rr} \frac{\partial c^{(\varkappa)}}{\partial r} + E_{r\theta} \frac{\partial c^{(\varkappa)}}{\partial \theta}\right), \tag{2}$$

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$$\prod_{\theta}^{(\varkappa)} = \varrho\left(\frac{c^{(\varkappa)}}{r} - E_{r\theta}\frac{\partial c^{(\varkappa)}}{\partial r} - E_{\theta\theta}\frac{\partial c^{(\nu)}}{\partial \theta}\right) \qquad (3)$$

are the isotope flow components along the axes; ϱ is the air density, $\lambda^{(\varkappa)}$ is the radioactive decay constant, σ is the aerosol washout coefficient ($\sigma = 0$ by $r > h(\theta)$), v_r, v_θ are the velocity components of zonally averaged meridional circulation, w is the gravitational sedimentation velocity of aerosols, and E_{rr}, $E_{r\theta}$, $E_{\theta\theta}$ are the components of macroturbulent diffusion tensor in the meridional plane. In the lower stratosphere the last components are connected by the relations

$$E_{\theta\theta} = K_\theta a^{-2}, \quad E_{r\theta} = \alpha K_\theta a^{-1}, \quad E_{rr} = K_z + \alpha^2 K_\theta \qquad (4)$$

with "normal" vertical K_z and meridional K_θ macroturbulent diffusion coefficients and with an angle of slope α of tensor main axe to the surface of the Earth with radius a (Reed & German, 1965; Karol, 1966). In the troposphere $\alpha = 0$ and therefore $E_{r\theta} = 0$, $E_{rr} = K_z$.

The following isotopes of the radon decay chain are considered: Rn^{222} ($\varkappa = 0$; $\lambda^{(0)} = 2.1\ 10^{-6}$ sec^{-1}) for which $\sigma = w = 0$, Pb^{210} ($\varkappa = 1$; $\lambda^{(1)} = 1.0 \cdot 10^{-9}$ sec^{-1}), Bi^{210} ($\varkappa = 2$; $\lambda^{(2)} = 1.6 \cdot 10^{-6}$ sec^{-1}), Po^{210} ($\varkappa = 3$; $\lambda^{(3)} = 5.8 \cdot 10^{-8}$ sec^{-1}) which are assumed to be connected whith the same aerosols, and therefore to have the same σ and w values.

The boundary conditions of the problem are:

$$\text{(a)}\ \prod_{\theta}^{(\varkappa)}\Big|_{\theta=0} = \prod_{\theta}^{(\varkappa)}\Big|_{\theta=\Pi} = 0, \quad r_1 < r < H; \qquad (5)$$

$$\text{(b)}\ \left[\prod_{r}^{(\varkappa)} + \varrho(w + \beta^{(\varkappa)})c^{(\varkappa)}\right]_{r=H} = \varrho\psi^{(\varkappa)}, \quad v_r\big|_{r=H} = 0 \qquad (6)$$

is the condition at the upper boundary $r = H$, which follows from the assumption that for $r > H$ only the vertical macroturbulent diffusion of considered isotopes takes place with the same coefficients as at level $r = H$. Here $\beta^{(\varkappa)}$ is the parameter introduced by the author (1966), $\psi^{(0)} = \psi^{(1)} = 0$, $\psi^{(\varkappa)} = b^{(\varkappa)}(\theta)\lambda^{(\varkappa-1)}$, $c^{(\varkappa-1)}(H, \theta)$, $\varkappa = 2.3$.

$$b^{(2)} = \frac{\beta^{(1)} - \beta^{(2)}}{\lambda^{(2)} - \lambda^{(1)}}, \quad b^{(3)} = \frac{\beta^{(2)} - \beta^{(3)}}{\lambda^{(3)} - \lambda^{(2)}} + \sum_{i=1}^{3} \frac{\beta^{(i)}\lambda^{(i)}}{\prod_{\substack{j=1 \\ j\neq i}}^{3} (\lambda^{(j)} - \lambda^{(i)})}, \qquad (7)$$

$$\text{(c)}\quad K_z \frac{\partial c^{(0)}}{\partial r}\bigg|_{r=r_1} = -\delta\varepsilon\varrho(r_1), \quad \left[K_z\frac{\partial c^{(\varkappa)}}{\partial r} - vc^{(\varkappa)}\right]_{r=r_1} = 0 \qquad (8)$$

are the conditions at the level r_1 of isotope concentration measurements in the ground level air ($r_1 = 1$ m usually), where ε is the radon exhalation rate, δ is the land area per cent in the zonal belt area, and v is the velocity of aerosol "dry deposition" on the ground.

To take into account almost the linear growth of $K_z = k_1 r$ in the lower part $r < L$ of the atmosphere boundary layer and to avoid the difficulties in finite difference approximation of equation (1) there the conditions (8) are transferred to the level L, preserving the $c^{(\varkappa)}$ and $\Pi_r^{(\varkappa)}$ continuity.

To obtain the numerical values of parameters and coefficients entering in the equations and boundary conditions the great number of climatic data on wind, temperature, and density distributions in the troposphere and lower stratosphere of both hemispheres is used.

The annually and zonally averaged v_θ in the northern hemisphere is taken (a) from the known Crutcher's atlas, presented by Holopainen (1967), for the troposphere, and from Peng (1963, 1965) for 100–30 mb layer, (b) from Guterman (1965) where the wind data over the USSR territory are presented more completely. In the southern hemisphere the calculated averaged v_θ of Gilman (1965) is used in both variants. The annually and zonally averaged v_r is calculated by numerical integration of the zonally averaged stationary continuity equation, using the real mean density data and the additional condition $v_r|_{r=H} = 0$. The obtained meridional circulation pattern has the known three cells in the troposphere of each hemisphere, with lower intensity of circulation in the southern hemisphere and greater intensity of the polar cell in variant b.

The annually and zonally averaged E_{rr}, $E_{r\theta}$, $E_{\theta\theta}$ in variant A are calculated using the climatic data on zonally averaged mean square $\overline{\nabla}_\theta$ deviations, due to transient and standing eddies from (Newell *et al.*, 1966; Peng, 1963, 1965; Obasi, 1963); α was assumed to be approximately equal to the angle of tropopause slope to the poles at all levels of corresponding latitude. The estimates of K_z in the stratosphere are

based on some results of Newell, (1963) and Karol (1966). The details of all these calculations will be published elsewhere and the resulting vertical profiles of E components are presented in Figs. 1 and 2 for several latitudes of both hemispheres.

In variant B for the stratosphere the annually averaged components obtained by Reed & German (1965) are used and for the troposphere K_θ and K_z are four times greater on the average than in variant A. E_{rr} is almost equal for A and B variants at the level $r = H$, but Reed & German's E_{rr} increase with decreasing r faster than those in A variant and in 12–16 km layer the former are 2–4 times greater than the latter. Almost the same relation holds for $E_{r\theta}$ and $E_{\theta\theta}$ in B and A variants. The method of w_1 estimation in the stratosphere is the same as that of Karol (1965, 1966).

As the Rn^{222} and Pb^{210} concentrations in the layer $r_1 < r < h$ of aerosol washout are 10–100 times higher over the land than over the ocean, then the long-lived radon daughter fallout takes place mainly over the land. Evidently the land values of aerosol removal parameters must be used in the equation (1) and boundary condition (8). So the following values are used: $L = 100$ m in A and 300 m in B variants on the average (twice lower in polar zones); $k_1 = 0.1$ (m²/sec) on the average (0.05 in the polar zone, 0.15 in the tropical zone); $\varepsilon = 0.7$ atom/cm² sec = 0.4 pc/m² sec the same in all latitudes, except polar zones, where $\varepsilon = 0$ at 80° N and to the south of 60° S and $\varepsilon/2$ and $\varepsilon/4$ at 60° and 70° N; δ is the same as this of Karol (1965); $v = 0.3$ cm/sec in A and 0.6 cm/sec in B variants, following Lambert & Nezami (1965), and Davidson *et al.* (1966) in all latitudes. σ is independent of height in the washout layer $r_1 < r < h$ just as it was taken by Karol (1965), where h changes from 2 km in polar regions to 4 km at the equator. σ is proportional to cli-

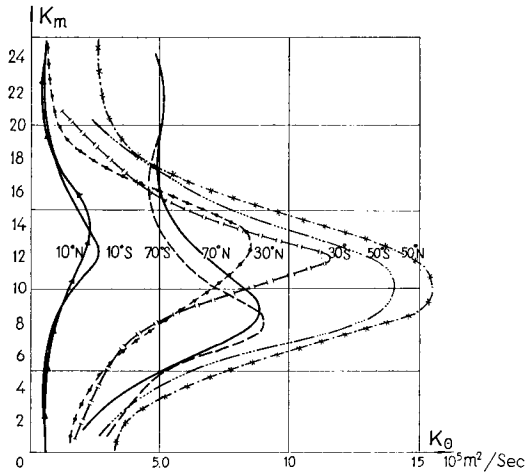

Fig. 1. Meridional macroturbulent diffusion coefficient K_θ (m²/sec) for variant *A*.

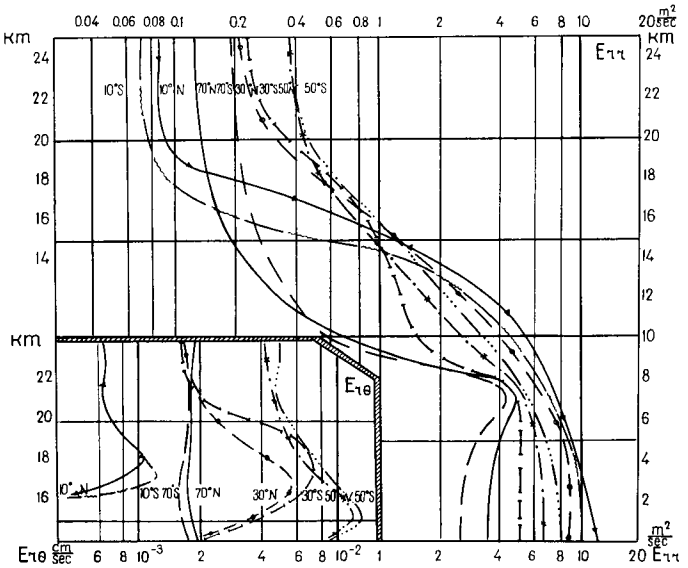

Fig. 2. Components E_{rr} (m²/sec) and $E_{r\theta}$ (cm/sec) of macroturbulent diffusion tensor for variant *A*.

Table 1. *Calculated variants*

No. of variant	1	2	3	3*a*	3*b*	4	5	6	6*a*	6*b*
E-values	A variant (case I)					B variant (case II)				
$v_r, v\,\overline{\nabla}_\theta$	0	a	b	b	b	0	b	a	a	a
w_i for r_i (μ)	0.1	0.1	0.1	0.0	0.5	0.1	0.1	0.1	0.1	0.1

matic annual precipitation amount over the land in the 10° latitudinal belt (Zubenok, 1956) and σ changes from $1.5\text{–}2.10^{-6}$ sec^{-1} at poles to $4\text{–}5.10^{-6}$ sec^{-1} in the tropical zone.

The equation (1) and boundary conditions (5)–(8) are approximated by their finite difference analogues with differences $\Delta\theta = 10° = 0.1745$ and $\Delta r = 1$ km ($19 \times 26 = 494$ grid points for the considered rectangle) and then solved numerically using the matrix factorisation method (Marchuk 1961) by an electronic computer. For verification of solution the balance equation is calculated: the sum of each isotope atom source and sink intensities in the rectangle and atom flows across its boundaries must be close to zero.

The flows of atoms across the equator and the tropopause are calculated together with the "dry" and "wet" fallout densities as latitude functions and their sums for the northern and southern hemispheres.

To investigate the influence of different atmospheric processes on isotope concentration patterns and flows of theirs atoms, several variants are calculated. Among them two cases are considered in detail here and appropriate parameters are presented in Table 1.

The patterns of calculated specific activity $\lambda^{(x)}c^{(x)}(r, \theta)$ isolines are presented for Pb^{210} and Po^{210} in Figs. 3 and 5 for case I (variants 1–3) and in Figs. 4 and 6 for case II (variants 4–6). The difference between the full ($\mathbf{v} = 0$), dashed ($\mathbf{v}$, variant a) and dotted-dashed ($\mathbf{v}$, variant b) isolines for all isotopes is more distinct in case I with weak intensity of macroturbulent diffusion than in case II with strong one. The positive v_r in the lower troposphere of equator, 60° N and 60° S zones increase

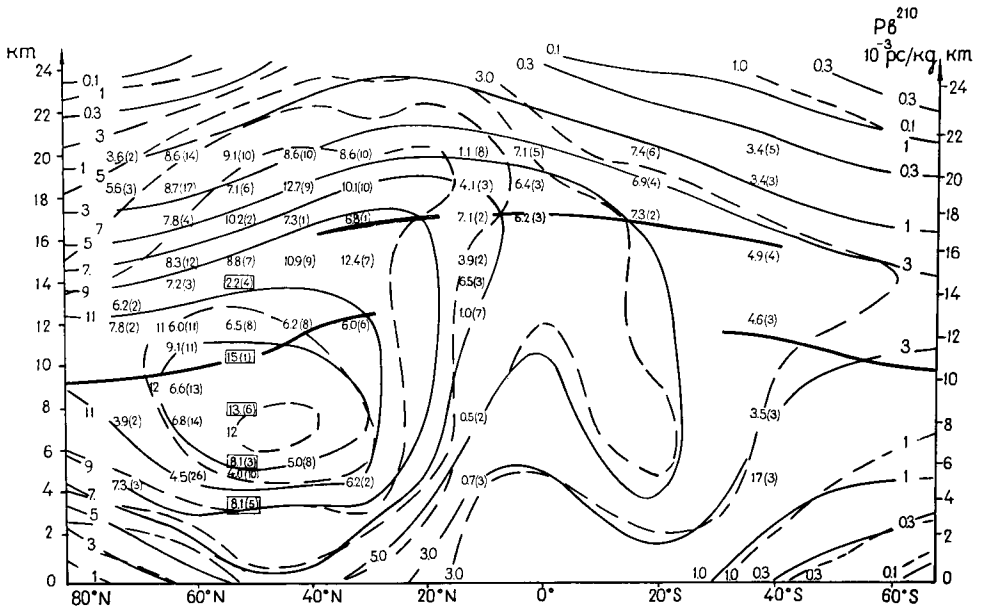

Fig. 3. Isolines of calculated Pb^{210} concentration for case 1: variant 1 (——) and variant 2 (– – –), and their average values measured in 1962–63. Numbers of averaged measurements are in brackets here and in the following figures. Data in frames were obtained over eastern Atlantic in 1954–56 (Burton & Steward, 1960).

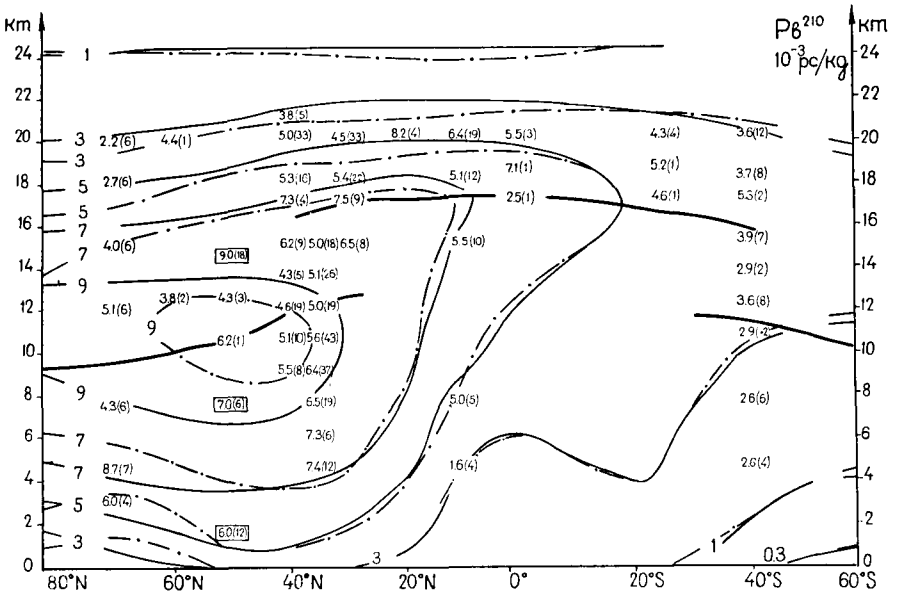

Fig. 4. Isolines of calculated Pb^{210} concentration for case II; variant 4 (——) and variant 5 (·—·), and their average values measured in 1960–61 and 1964–65. Data in frames were obtained over eastern Atlantic in 1958 (Peirson *et al.*, 1966).

(negative in 20–30° N and 10–20° S zones decrease) the concentration of Rn^{222} (ground level source) at these latitudes, but for Pb^{210} and Po^{210} (upper troposphere and lower stratosphere sources) the same v_r lead to the opposite concentration change in the lower troposphere. The increased isoline slope for all isotopes in case I compared to case II in the 18–25 km layer is due to decrease with height of main axis slope angle α of diffusion tensor E in Reed &

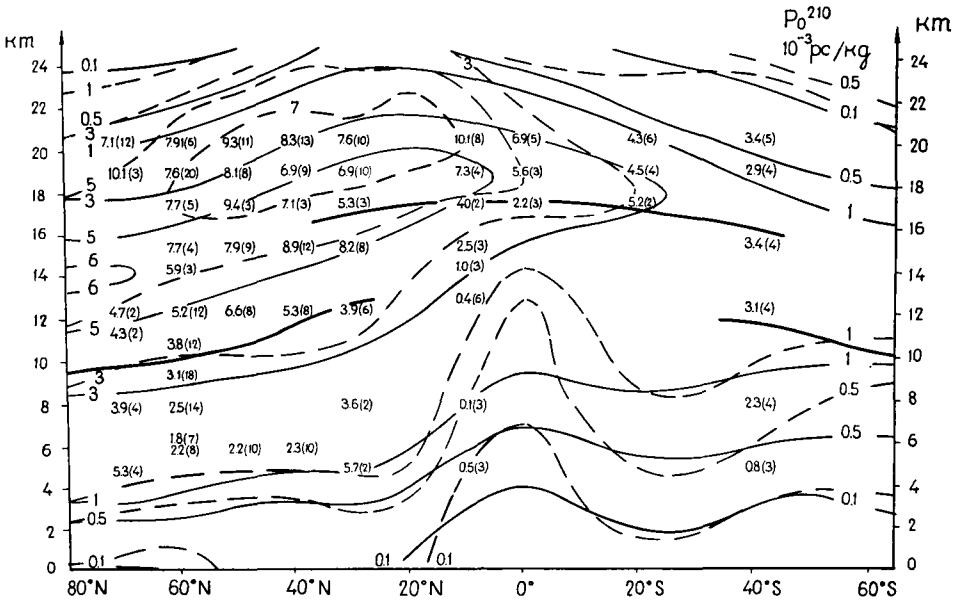

Fig. 5. As Fig. 3, but for Po^{210}.

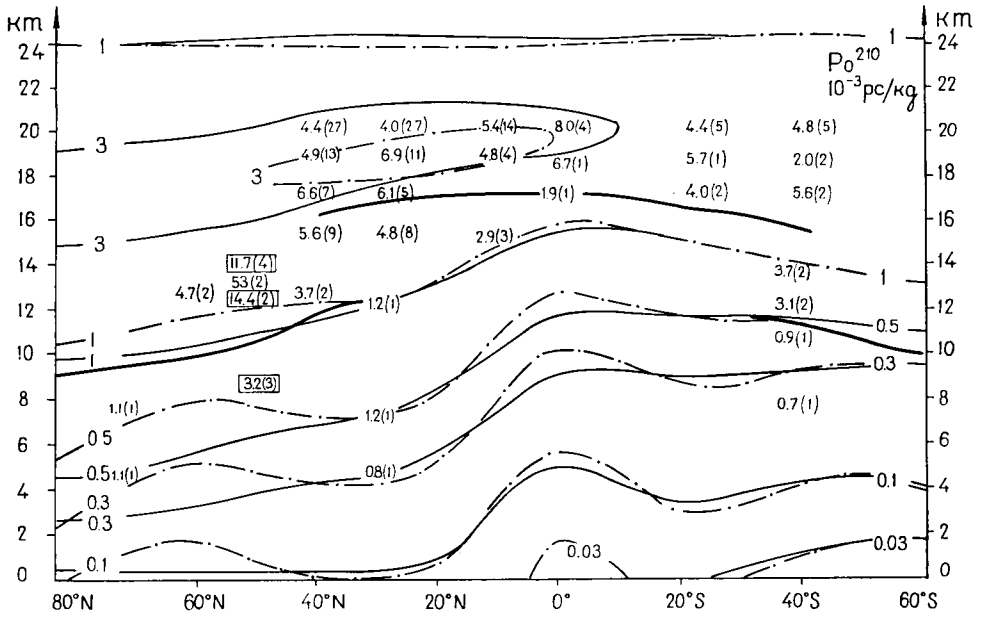

Fig. 6. As Fig. 4, but for Po^{210}.

German's model (case II) compared to constancy of α with height in case I. The significant increase of stratospheric Po^{210} concentration in variants 2 and 3 (case I) compared to case II is due to diminished diffusion intensity and enhanced Pb^{210} transport in this layer by ordered motions. The radioactive equilibrium between Pb^{210} and Bi^{210} ($\lambda^{(1)}c^{(1)} \simeq \lambda^{(2)}c^{(2)}$ with the first two significant figures) takes place in all the stratosphere in case I and for $r > 16$–19 km in case II, but the same equilibrium between Pb^{210} and Po^{210} ($\lambda^{(1)}c^{(1)} \simeq \lambda^{(3)}c^{(3)}$) is reached only for $r > 22$ km in tropical zone and for $r > 20$–22 km outside it in case I and it is not established at all up to 25 km in case II.

Comparing the calculated here and measured isotope specific activity in the atmosphere one must bear in mind that the latter depend strongly on position of measurement place relative to continents and oceans. Therefore the deviation of measured from calculated zonally averaged concentration can be significant especially in the troposphere. The Pb^{210} and Po^{210} specific activity data were obtained by the aircraft measurements at 70° N over Alaska; in the temperate latitudes—over the western and central part of North America and over the eastern Atlantic, in tropics—over the western and central Pacific and Central America; in the southern hemisphere—over the eastern Australia, and published by Burton & Stewart (1960), Peirson *et al.* (1966), Machta (1965), HASL USAEC (1956, 1966). These data are averaged for years 1960–61 and 1964–65 (period I) and for 1962–63 (period II) and presented by numbers in the Figs. 3 and 5 and 4 and 6, respectively. The division of data into two periods is connected with the possibility proposed by Machta (1965) & Peirson *et al.* (1966), that some quantity of Pb^{210} was produced in 1961–62 nuclear test explosions. The comparison of data presented in figures shows that not only Pb^{210} but also Po^{210} concentrations in period II is 1.5–2 times higher than in period I, especially in 18–20 km layer.

The agreement between measured and calculated Pb^{210} activities is satisfactory in both cases. Calculated values of Pb^{210} specific activity exceed measured ones in the northern hemisphere temperate zone probably due to the influence of the Pacific in the upper troposphere over the western part of the United States, where the measurements were performed. But calculated Po^{210} activity agrees with measured one only in variants 2 and 3 of case I, and is apparently lower than the data in case II at all levels. This discrepancy is most probably due to the overestimation of macro-

Table 2. *Isotope content (10^{22} atoms) and their fallout (10^{22} atoms/year)*

Isotope	Rn^{222}			Pb^{210}			Po^{210}		
No. of variant...	1	2	5	1	2	5	1	2	5
N. hemisph. troposph.	25.7	25.9	24.1	58.5	54.7	43.4	0.161	0.139	0.048
S. hemisph. troposph.	10.8	10.6	10.2	23.2	22.0	17.8	0.076	0.060	0.025
N. hemisph. stratosph.	7.10^{-3}	9.10^{-3}	0.15	9.4	11.4	7.9	0.090	0.115	0.042
S. hemisph. stratosph.	8.10^{-4}	6.10^{-4}	0.03	3.4	3.6	4.8	0.037	0.042	0.037
Total	36.5	36.5	34.5	94.5	90.7	73.9	0.364	0.356	0.152
N. hemisph. fallout		1830[a]		1740	1740	1640	1.04	0.99	0.85
S. hemisph. fallout		740[a]		720	710	700	0.26	0.22	0.28
Total		2570[a]		2460	2450	2340	1.30	1.21	1.13

[a] Integrated exhalation from land surface.

turbulent diffusion intensity at all levels in case II, which is not so evident for Pb^{210}. The Po^{210} serves better than Pb^{210} for stratospheric global exchange investigation as its time scale is comparable with Po^{210} half-life (138 days) and is much lower than Pb^{210} half-life (20 years). Thus the lower intensity of macroturbulent diffusion in case I is closer to reality and the ordered meridional circulation has the prominent influence upon the isotope concentration distribution.

The Pb^{210} specific activity values calculated in variants 3 a) and 3 b for other gravitational settling velocities w_1 differ significantly from those in variant 3 (Fig. 3) higher than 15 km. At 20 km Pb^{210} activity is 2–3 times higher for $w_1 = 0$ (variant 3 a) and 5–20 times lower for w_1 corresponding to $r_1 = 0.5\ \mu$ (variant 3 b). Comparing this with observational data one can come to the conclusion that $r_1 = 0.1\ \mu$ is the most probable "critical value" (Karol, 1966) of natural radioactive aerosol radius in the lower stratosphere, that agrees well with results of Martell (1966).

The calculated zonally averaged Pb^{210} fallout agrees well with the estimates of Lambert & Nezami (1965) especially for variant 2 in the Northern temperate latitudes and in the Antarctic, where the estimates are based on direct measurements. The agreement is worse in the tropics and southern temperate latitudes, where the estimates were obtained by an indirect way. The calculated part of "dry" deposition changes from 20–30 % for case I and 6 a variant of case II with 0.3 cm/sec to 30–40 % for variants 4–6 of case II, that is lower than 50 % estimate (Lambert & Nezami, 1965).

The calculated ratio of Bi^{210}/Pb^{210} specific activities in rain and ground level air of northern temperate latitudes changes from 0.35–0.40 for variants 1–3 and 4–6 to 0.45–0.50 for variant 6a, that is close to few measurements of this ratio (Karol & Vilenskii, 1965). But the analogous Po^{210}/Pb^{210} ratio for all variants does not exceed 0.03–0.05, which is much lower than average measured values 0.10–0.15 (Burton & Stewart, 1960; Peirson *et al.*, 1966; Lambert & Nezami, 1965). The Po^{210}/Pb^{210} calculated ratio attains the latter values in ground level air (but not in precipitation) only in variant 6[b] when the aerosol removal parameters σ and v for Po^{210} are arbitrary taken 3 times lower than for other isotopes. The reasons of this discrepancy between calculated and measured values are yet unclear.

The calculated content of Rn^{222}, Pb^{210} and Po^{210} atoms in the troposphere and stratosphere and their fallout in the northern and southern hemispheres are presented in Table 2. The ratio of Rn^{222} and Pb^{210} atom content in the northern hemisphere to the southern hemisphere (2.4–2.6) approximately coincides with this ratio for integral Rn^{222} exhalation (2.5). The same is true for Pb^{210} fallout.

The isotope content in the troposphere for Pb^{210} is 5–7 times and for Po^{210} 1.2–2 times higher than in the stratosphere of each hemisphere for variants 1 and 2 and in the northern hemisphere for variant 5. About 95 % (97 %) of Rn^{22} atoms exhaled into the atmosphere from the land surface of the northern (southern) hemisphere fall as Pb^{210} atoms on the surface of the same hemisphere. Only about 0.06 % (variant 1), 0.9 % (variant 2), 0.6 % (variant 3)

of Pb^{210} atoms annually formed in the atmosphere of northern hemisphere are transferred into the southern hemisphere (20 % as estimated by Lambert & Nezami (1965)). So the Pb^{210} transport across the equator is poor and is performed principally by meridional circulation (which is absent in variant 1).

The integrated flows of isotopes across the tropopause (which are not presented here for the sake of brevity) have almost the same order of magnitude for vertical component and horizontal one across the subtropical tropopause break (excluding the Rn^{222} in the northern hemisphere). In both hemispheres these flow components transport Pb^{210} between troposphere and stratosphere in opposite directions (e.g. in the northern hemisphere the vertical integrated flow is directed into the stratosphere, horizontal one out of it). That makes difficult the exact determination of net isotope transport across the tropopause, but shows clearly that the subtropical tropopause break is the significant channel for it. The Rn^{222} and Pb^{210} net transport across the tropopause and their "residence time" in the stratosphere of each hemisphere for the case I have the same order of magnitude as their previous estimates by Karol (1965).

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РАСЧЕТ ПЛАНЕТАРНЫХ РАСПРЕДЕЛЕНИИ КОНЦЕНТРАЦИЙ РАДОНА И ДОЛГОЖИВУЩИХ ПРОДУКТОВ ЕГО РАСПАДА В ТРОПОСФЕРЕ И НИЖНЕЙ СТРАТОСФЕРЕ

Рассматривается краевая задача для уравнения в частных производных для зонально усредненной концентрации трассера в меридиональной плоскости до уровня 25 км и от полюса в связи с некоторой моделью процессов переноса примеси в атмосфере в глобальном масштабе. Коэффициенты этого уравнения определяются из климатических данных по меридиональной циркуляции и макротурбулентной диффузии. Конечноразностный аналог этой задачи решается численно методом матричной факторизации и получены среднегодовые, зонально-усредненные распределения концентрации радона, свинца-210, висмута-210 и полония-210 для ряда вариантов параметров.

Результаты расчета сравниваются с опубликованными данными измерений концентрации изотопов в свободной атмосфере и оценивается глобальная интенсивность макротурбулентной диффузии. Кратко упомянуты результаты расчета суммарного содержания изотопов в тропосфере и стратосфере северного и южного полушария и скорости их переноса через экватор и через тропопаузу.