The size distribution of radioactive particles from nuclear weapon tests and their transport in the atmosphere

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(Manuscript received October 12, 1965)

ABSTRACT

In 1964–1965 in the Moscow region radioactive particle size distribution was studied in the ground layer of the atmosphere and in December, 1964, measurements were carried out at 5–6 km as well. Four kinds of radioactive aerosol size distribution variations were observed. In the troposphere at 5–6 km radioactive particles were on the average smaller than in the ground layer. The mean diameter of particles-carriers of Cs^{137} at 5–6 km was greater than that of particles-carriers of Ce^{144}. This difference was smoothed in the samples, taken at the ground surface.

Analysis of literature data on variations of ratios of various isotope concentrations was made. It shows that radioactive particle size distribution significantly effects the transport of these particles in the atmosphere, especially in the higher layers of the stratosphere and in the mesosphere.

Introduction

At the present time the most important components of atmospheric aerosols are artificial radioactive aerosols formed as the result of weapon tests in the atmosphere. (Further in this report for short aerosols from nuclear explosions will be referred to as radioactive aerosols or simply aerosols. Natural radioactive aerosols will not be considered at all in this report.) The physical properties of these aerosols and the character of their distribution in the atmosphere are not yet studied thoroughly. In particular there are very few data on aerosol size distribution of global radioactive atmospheric contamination, on isotope fractionation with particle size and peculiarities of transport of various aerosol fractions in the atmosphere. But these data are of considerable value for an understanding of aerosol particle behaviour in the atmosphere. Therefore, in recent years in the Moscow region we studied aerosol particle size distribution, fractionation of Ce^{144}, Cs^{137}, Ru^{106} and Mn^{54} with particle size and relations between aerosol particle size distribution and their transport during the period of moratorium. In this report the results obtained will be discussed. Preliminary results of our research were reported earlier (GASIEV *et al.*, 1965*a*) at the conference on nuclear meteorology held in February, 1964, in Obninsk, USSR.

Technique of measurements

For the measurements discussed in this paper two kinds of samples were used both at the ground level and in the aircraft.

1. Cascade impactors with the following characteristics. Theoretically and experimentally roughly estimated size range for the particle diameters on the various stages if the density of particles is assumed to be $\varrho = 1$ g/cm³:

1. Stage $d > 2.5\ \mu$
2. Stage $1.0 \leqslant d < 2.5\ \mu$
3. Stage $0.3 \leqslant d < 1.0\ \mu$
4. On the fine filter (of type ФПП-15-1.5, OGORODNIKOV *et al.* 1965) $d < 0.3\ \mu$

Sampling rate at the ground level was 38 m³/h and at 5000–6000 m altitude in the aircraft was 80 m³/h.

2. Impactor-filter devices consisting of a synthetic grid and a fine filter (ФПП-15-1.5). In the device used at the ground level the synthetic grid is followed by the filter, in the aircraft device the grid is used parallel with the filter. The diameter of the grid threads is about 50 μ and very uniform.

The collection efficiency E for the synthetic grid is shown in Fig. 1. The values of E_1 at the ground level are taken from GASIEV *et al.*, 1965*a* and refer to a face velocity of about $u = 50$ m/sec. The values of E_2 are equal to those of E_1 but corrected for sampling conditions in the aircraft, i.e. for $u = 40$ m/sec, air pressure $p = 380$ m Hg and air temperature $t \approx 30°C$. Fig. 1 shows that most of the particles with $d > 0.65$ μ are collected by the grid. Most of the particles with $d < 0.45$ μ pass through the grid. To assure collection on the grid it was covered by a thin layer of sticky material.

The sampling rate for the devices used at the ground level was 230 m³/h and for the devices used in the aircraft 7000 m³/h for the grid and 800 m³/h for the filter. The impactor-filter device was covered when the aircraft flew in the cloud layers. On the ground during precipitations a box was placed near the sampling device. This box had two openings one of which was connected with the inlet of the device and the other, very large, was covered by gauze. The rate of air pumping through the opening covered by gauze was very low. Besides, this sampling device and the box were placed under a shed. Therefore the collection of rain drops and snow flakes by the grid and filter was prevented.

The area of the threads is about 35% of the total grid area. The radioactivity of particles deposited on the threads of the grid, therefore, refers to 35% of the air volume drawn through the grid. The percent fraction, α, of the radioactivity attached to the large particles is thus for the ground level device

$$\alpha = 100\ A_1/0.35\ (A_1 + A_2),$$

and for the aircraft device

$$\alpha = 100\ A_1 V_2/0.35\ A_2 V_1,$$

where A_1 = β-activity of the grid sample, A_2 = β-activity of the filter sample, V_1 = air volume drawn through the grid, and V_2 = air volume drawn through the filter.

The β-activity was determined by ashing the samples at temperatures of 500–600°C and counting them by Geiger-Muller counters of the type MCT-17* and CTC-5 with backgrounds of 10–11 counts/min and 1 count/min, respectively. γ-spectra were examined with the help of a 100-channel scintillation spectrometer with

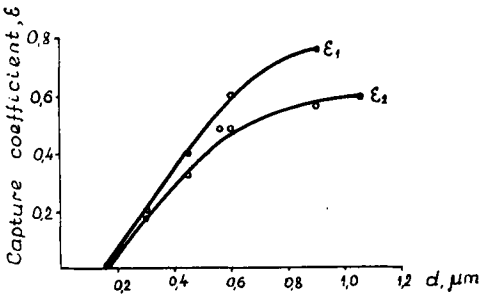

FIG. 1. Capture coefficients of aerosol particles by synthetic grid the impactor-filter device. ε_1 for the ground device; ε_2 for the aircraft device.

Na 1 (Tl) crystal the height and the diameter of which is 4 cm. To diminish the background the detecting element of the spectrometer was shielded. The wall thickness of the shield was 20 cm. Isotopic activity was given to one and the same date 1.01.1965.

Particle size distribution and fractionation of Ce^{144}, Cs^{137}, Ru^{106} and Mn^{54} with particle size

Measurements of particle size distribution in the surface layer of the atmosphere were started in January, 1964, with the help of an impactor-filter device and in April, 1964, with the help of a cascade impactor. Fig. 2 shows the mean monthly β-activities of radioactive particles of the 1st to 4th fractions retained by the cascade impactor expressed in percent of the total β-activity. Samples were taken daily in the day-time during 8–12 hours. In Fig. 2*a* it is seen that β-activity concentration in the air referring to the particles with $d < 0.3$ μ remained approximately constant ($\sim 40\%$ from April through June, 1964). Then it gradually decreased down to 3% (December, 1964). During the next months this relative concentration increased passing the maximum (about 32%) in April, 1965. Quite another picture presents itself with the 2nd and 3rd fractions. They have maxima in winter. Considerable fluctuations of average monthly relative β-activity concentrations are observed.

From the measurements carried out it was deduced that the most part of radioactivity (about 90%) was attached to the particles having diameters less than 2.5 μ. It is interest-

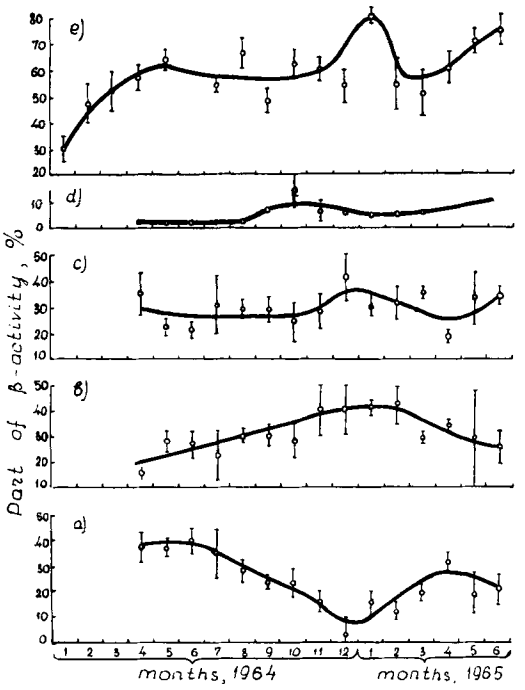

FIG. 2. β-activity distribution with particle size in the ground layer vs. time. *a*, *b*, *c*, *d*, activity retained by the 4th, 3rd, 2nd and 1st stages of the cascade impactor, respectively; *e*, β-activity relative concentration of large particle fraction from measurement data obtained by impactor-filter device.

ing to note that from November, 1964, through March, 1965, about 80% of radioactivity were on particles having diameters from 0.3 μ to 2.5 μ, i.e. in this period the radioactive particles size distribution was relatively narrow.

It should be noted that there is a general tendency to the decrease of the part of β-activity belonging to smaller particles during the period from April, 1964, through June, 1965, and to the corresponding increase of the part of β-activity belonging to large particle fraction, though this tendency is not very pronounced. Mean monthly relative concentrations of large particle fraction (α monthly) retained by synthetic grid of the impactor-filter device are shown in Fig. 2. From here the winter maximum and the general tendency to the increase of the part of large particle fraction (α monthly) from January, 1964, through June, 1965, are seen, though not very well. Fluctuations of α monthly values are also well seen. Similar α-fluctuations are especially well marked when averaging over the smaller period of time (1–3 days) (GASIEV, 1965*a*; LOCKHART *et al.*, 1965). Fluctuations observed from January, 1964, through June, 1965, are given in Fig. 3.

In December, 1964, aircraft investigations of radioactive particle size distribution were also

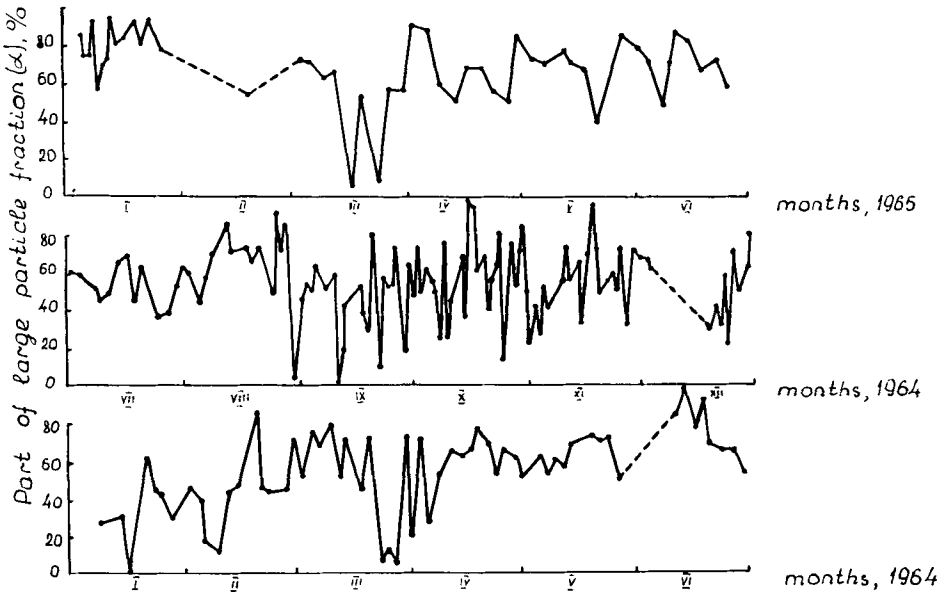

FIG. 3. Time variations of relative content of large radioactive particle fraction in the ground layer of the atmosphere.

TABLE 1. *Radioactive particle size distribution at the altitudes 5–6 km in December, 1964.*

Sampling date 1964	Relative part of β-activity			
	1 fraction $d > 2.5\,\mu$	2 fraction $1 < d < 2.5\,\mu$	3 fraction $0.3 < d < 1\,\mu$	4 fraction $d < 0.3\,\mu$
12–13.XII	7	14	23	56
14–15.XII	5	29	31	35
16–18.XII	3	26	27	44
18–22.XII	1	25	19	55
26–28.XII	7	23	20	50
Average	5	23	24	48
Standard deviation of the mean	1.2	2.5	2	3.5
Standard deviation of a single measurement	2.6	5.7	4.5	7.8

carried out in the middle troposphere at altitudes of 5–6 km.

The data obtained by the aircraft cascade impactor are given in Table 1.

It is seen from Table 1, that in December, 1964, at an altitude of 5–6 km on the average about 48% of β-radioactivity were on the particles with the diameter $d < 0.3\ \mu$ while in the ground layer only 3% of β-activity were found on the corresponding fraction. In November, 1964, and in January, 1965, the difference in radioactivity belonging to the fraction denoted above between the surface layer and the middle troposphere ($\sim 16\%$ and $\sim 48\%$, respectively) was somewhat lower.

To determine the pecularities of isotope distribution with particle size samples obtained by impactor-filter devices were analysed by γ-spectrometer. γ-spectrometer data are given in Tables 2–4.

Table 2 shows that there is a considerable difference in Ru^{106} and Ce^{144} distribution with particle size, large particle fraction being depleted with Ru^{106} and rich in Ce^{144}. As for Cs^{137} and Mn^{54}, they are distributed approximately equally between both fractions.

Unfortunately, a similar table could not be made for the samples collected in the middle troposphere because of large errors in determination which were due to great inaccuracy

TABLE 2. *Isotopic fractionation with particle size in the ground layer.*

l, large particle fraction; f, fine particle fraction.

Samling date 1964	Isotope part in size fractions, %							
	Ce^{144}		Cs^{136}		Ru^{106}		Mn^{54}	
	l	f	l	f	l	f	l	f
31.III–30.IV	50	50	49	51	20	80	44	56
4.V–1.VI	73	27	41	59	45	55	66	34
30.VI–31.VII	55	45	50	50	44	56	41	59
1.VIII–31.VIII	72	28	64	36	36	64	55	45
1.IX–30.XI	62	38	59	41	30	70	57	43
Average	62	38	53	47	35	65	53	47
Standard deviation of the mean	4.5	4.5	4	4	4.6	4.6	4.5	4.5
Standard deviation of a single measurement	10	10	9	9	10.4	10.4	10	10

TABLE 3. *Isotopic ratios in aerosol fractions.*
l, large particle fraction; t, total sample.

Sampling date 1964	Samling altitude (g = ground layer)	Isotopic ratios Ce^{144}/Cs^{137} l	Ce^{144}/Cs^{137} t	Ce^{144}/Mn^{54} l	Ce^{144}/Mn^{54} t	Ce^{144}/Ru^{106} l	Ce^{144}/Ru^{106} t
31.III–30.IV	g	6.8	6.6	13.2	11.3	5.8	2.1
4.V.–1.VI	g	7.7	3.6	14.0	12.0	3.5	1.9
30.VI–31.VII	g	5.5	4.4	15.5	11.0	2.9	2.3
1.VIII–31.VIII	g	6.4	4.8	17.0	11.8	6.0	2.5
1.IX–30.XI	g	6.4	6.0	12.9	11.7	5.9	2.4
Average		6.6	5.1	14.5	11.6	4.8	2.3
Standard deviation of the mean		0.4	0.54	1.3	0.2	0.7	0.15
Standard deviation of a single ratio		0.8	1.2	2.9	0.4	1.5	0.25
11.XII–13.XII	5000 m	3.1	7.4	10.7	7.3	2.3	2.6
14.XII–17.XII	5000 m	3.3	4.2	11.1	9.2	2.2	2.5
18.XII	5000 m	3.6	4.0	9.6	10.3	2.3	—[a]
19.XII–22.XII	5000 m	3.5	5.1	10.8	7.8	2.7	2.8
24.XII–26.XII	5000 m	3.4	4.3	11.4	12	2.5	2.7
Average		3.4	4.7	10.7	9.3	2.4	2.9
Standard deviation of the mean		0.1	0.6	0.3	0.8	0.1	0.03
Standard deviation of a single ratio		0.2	1.5	0.7	1.9	0.2	0.3

[a] Ru^{106} was not determined.

TABLE 4. *Isotopic fractionation.*
l, large particle fraction; t, total sample.

Sampling date 1964	Sampling altitude (g = ground layer)	$\frac{(Ce^{144}/Cs^{137})_t}{(Ce^{144}/Cs^{137})_l}$	$\frac{(Ce^{144}/Mn^{54})_t}{(Ce^{144}/Mn^{54})_l}$	$\frac{(Ce^{144}/Ru^{106})_t}{(Ce^{144}/Ru^{106})_l}$
31.III–30.IV	g	1.0	0.9	0.4
4.V–1.VI	g	0.5	0.9	0.5
30.VI–31.VII	g	0.8	0.7	0.8
1.VIII–31.VIII	g	0.8	0.7	0.4
1.IX–30.XI	g	0.9	0.9	0.4
Average		0.8	0.8	0.5
Standard deviation of the mean		0.1	0.05	0.1
Standard deviation of a single ratio		0.2	0.1	0.2
11.XII–13.XII	5000 m	2.4	0.7	1.1
14.XII–17.XII	5000 m	1.3	0.8	1.1
18.XII	5000 m	1.1	1.1	—[a]
19.XII–22.XII	5000 m	1.5	0.7	1.0
24.XII–26.XII	5000 m	1.3	1.1	1.1
Average		1.5	0.9	1.1
Standard deviation of the mean		0.2	0.1	0.03
Standard deviation of a single ratio		0.5	0.2	0.06

[a] Ru^{106} was not determined.

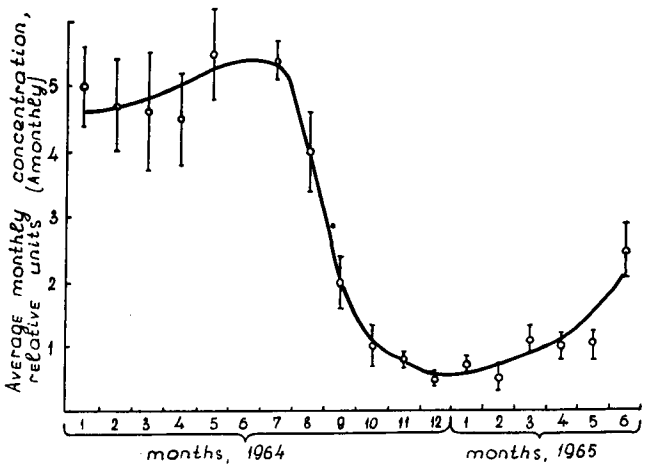

FIG. 4. Average monthly concentration of total β-activity in the ground layer of the atmosphere.

of measurements of air volume (V_3) drawn through the filter of the aircraft impactor-filter device.

γ-spectrometer data are given in the Tables 3 and 4. From Table 4 it is seen that in the troposphere at an altitude of 5 km the quotient of two ratios, Ce^{144} to Cs^{137} with index t and Ce^{144} to Cs^{137} with index l, is 1.5 ± 0.2. Here index t refers to the integral sample and index l to the large particle fraction. Thus, at an altitude of 5–6 km Ce^{144} is mainly connected with fine particles and Cs^{137} with large particles. In other words, the mean diameter of aerosol-carrier particles of Cs^{137} ($\bar{d}_{Cs}$) is greater than that of Ce^{144} ($\bar{d}_{Ce}$). The remaining two ratios are close to 1, that is, in the middle troposphere Ce^{144}, Ru^{106} and Mn^{54} are distributed with aerosol fractions equally. In the surface layer of the atmosphere the quotient of two ratios, Ce^{144} to Cs^{137} with index t and Ce^{144} to Cs^{137} with index l, is a little less than 1. Probably, because of the increased coagulation of radioactive particles with those of atmospheric dust, differences in distribution of Ce^{144} and Cs^{137} with particle size observed in the middle troposphere were smoothed. The quotient of two ratios Ce^{144} to Ru^{106} with index t and Ce^{144} to Ru^{106} with index l which is equal to 0.5 ± 0.1, shows that in the surface layer of the atmosphere the amount of Ru^{106} connected with large particles was relatively greater than that of Ce^{144}.

Experimental data analysis

Time variations of radioactive particle size distribution were studied very little. But as for peculiarities of seasonal variations of fission product concentrations in the ground layer of the atmosphere they were studied well and the main reasons responsible for these peculiarities were cleared up. Therefore it seems reasonable first of all to compare data on time variations of concentrations and radioactive particle size distribution. Concentrations of gross β-activity which were obtained in the Moscow region from January, 1965, are given in Fig. 4. Autumn-winter minimum and spring-summer maxima of concentrations are well seen here. As it is known the latter are due to the increase of radioactive particle flux from the stratosphere into the troposphere and the former is connected with its decrease.

Comparing Fig. 2 and Fig. 4 it is suggested that less influx of stratospheric aerosols is related to a smaller contribution of the small radioactive particle fraction (the 4th fraction, Fig. 2) into the gross β-activity of aerosols. During the periods of high β-activity concentration (higher influx of stratospheric aerosols), on the contrary, this contribution increases.

Apparently, radioactive particle size distribution in the ground layer of the atmosphere depends on the size spectrum of stratospheric radioactive particles, flux rate of these particles into the troposphere, intensity and duration of

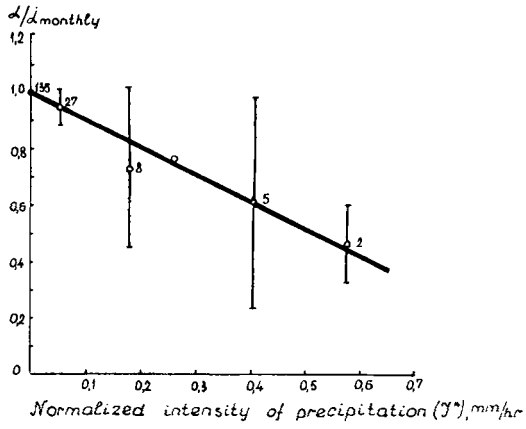

FIG. 5. Washout influence on α deviations from average monthly values.

the processes responsible for variation of particle size spectrum in the troposphere and exchange rate of air masses between the upper and lower troposphere. To all appearances, decrease of aerosol flux from the stratosphere into the troposphere from July through December 1964 (Fig. 4) under the conditions of existence of radioactive particle coagulation with natural tropospheric aerosols led to the gradual decrease of β-activity which was connected with the finest radioactive particle fraction in the ground layer of the atmosphere (Fig. 2*a*). Simultaneously the part of β-activity connected with large particle fraction increased because of enlargement of radioactive particles owing to coagulation with non-radioactive ones (Fig. 2). As it is seen from the difference of particle size distribution in December in the ground layer of the atmosphere and at altitudes of 5–6 km (Table 1) the most rapid enlargement of radioactive particles occurs, apparently, in the lowest layers of the atmosphere. Coagulation possibly increases additionally in winter months because of higher air pollution which is also suggested in the paper by LOCKHART *et al.* (1965). Increase of β-activity connected with the fraction of $d < 0.3$ μ from January, 1965, through June, 1965, probably depends on increasing the flux of stratospheric aerosols into the troposphere resulting in a shift of radioactivity to smaller particles. The fact was already mentioned that the main part of radioactivity of stratospheric aerosols is apparently attached to the particles less than 0.3 μ in diameter (DREVINSKY *et al.*, 1959; GASIEV *et al.*, 1965*b*). A general tendency to the increase of radioactive particle mean sizes from January, 1964, through June, 1965, in the ground layer of the atmosphere is probably the result of these radioactive particle coagulations with natural aerosols.

Coagulation and variation of size distribution of radioactive stratospheric particles after they appeared in the troposphere can take place because of difference in washout of various size particles (GREENFIELD, 1957). The ratio of α-value to average-monthly value of α vs. normalized precipitation intensity is given in Fig. 5. Normalized precipitation intensity is determined as $Y^* = q/\Delta t$, where q the whole amount of precipitation in mm, Δt duration of sampling in hours. If the duration of precipitations was less than $\Delta t/2$, then the value of α was not used for the analysis. From Fig. 5 it can be concluded that in the period considered the large particle fraction was on the average washed out better than the fine particle one, though the differences in washout rate of both fractions were not very large.

Decrease of relative concentration of the large radioactive particle fraction (α) can be due not only to washout. Under favourable meteorological conditions it can also be due to rapid flux of air masses into the ground layer of the atmosphere from the upper layers, especially from the upper troposphere and the lower stratosphere where sizes of radioactive particles are finer. However, in the first case α-decrease must be accompanied by the decrease of gross β-activity concentration in the air (A) and in the second case, on the contrary, this concentration increases. Correlations between α and β-activity values from January through May, 1964, confirming the suppositions given above were considered earlier (GASIEV *et al.*, 1965*a*). Relations between α/α monthly and A/A monthly, where α monthly and A monthly are average monthly values of α and A, are given in Fig. 6 for the period from January, 1964, through June, 1965. These data are consistent with the results obtained earlier. Cases when little α/α monthly and large A/A monthly are observed are given in Table 5.

Analysis of meteorological situation showed that in three of these cases (Nos. 1, 2 and 6) on sampling days or the days before sampling the troughs with sharp profiles were observed

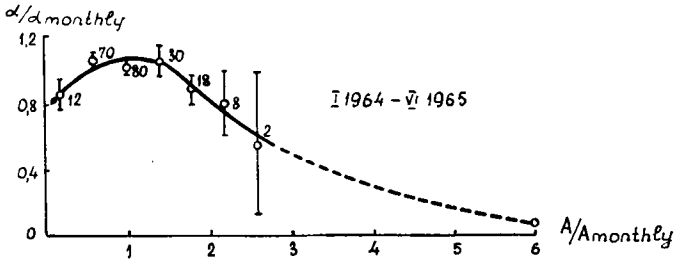

FIG. 6. Relations between α and A deviations from average monthly values. *Note:* In Figs. 5 and 6 a number of cases is marked by figures near points.

at high altitudes above the measurement area, and under the troughs at the ground surface in the cyclon rear the pressure increased. According to the works by DMITRIEVA (1965) and DIMITRIEVA & KASATKINA (1965) in these cases rapid air flux must take place from the stratosphere into the lower layers of the troposphere. For example the processes were not intensive enough on 9–11 September, 1964, but they were intensive from 15 through 18 January, 1964. In January (15–18) they were rather great at trough periphery and the tropopause descended down to 6–7 km. Then the cut-off cyclone formed. Pressure increase was observed under the trough at the ground surface in the cyclone rear. According to the work by DMITRIEVA & KASATKINA (1965) these conditions are especially favourable for radioactive aerosol entrance from the stratosphere into the ground layer of the atmosphere. As it is seen from Table 5, in this case and A deviations from average monthly values were extremely great. In other cases (Nos. 3, 4, 5) listed in Table 5, a similar meteorological situation above the measurement area was not observed. Probably this was due to the fact that aerosol flux from the stratosphere into the troposphere took place in the area which was situated far from the measurement points. The fact that α variations with time have fluctuating character is due to dependence of α values on a great number of factors variable with time, for example washout, coagulation, vertical and horizontal exchange of air masses (Fig. 3). Further accumulation of experimental α data and their subsequent statistical treatment with the account of the meteorological situation must make possible a better understanding of atmospheric processes changing particle size distribution in the ground layer of the atmosphere.

TABLE 5.

Sample number	Sampling data 1964	α/α monthly	A/A monthly
1	16–18.I	0.03	6.0
2	11.IX	0.30	2.6
3	7.X	0.40	2.2
4	9.X	0.40	2.4
5	24.X	0.20	2.2
6	22–23.III	0.20	1.8

The relation $d_{Ce} < d_{Cs}$ obtained by us during measurements at altitudes of 5–6 km in December, 1964, was also observed in 1961 in the stratosphere (GASIEV *et al.*, 1965). It is not impossible that in 1964 above the tropopause in the middle latitudes of the European territory of the USSR there were radioactive aerosols with $d_{Ce} < d_{Cs}$ which then entered the middle troposphere and were responsible for the observed Ce^{144} and Cs^{137} isotopic fractionation with radioactive particle size. It can be assumed that d_{Ce}/d_{Cs} took place because of stronger coagulation of Cs^{137} radioactive particles with aerosol particles with aerosol particles of Junge's layer in the stratosphere as the original (after explosion) particle sizes with which Cs^{137} is mainly connected are smaller than the original sizes of particles to which Ce^{144} is mainly attached (EDVARSON *et al.*, 1959). The causes of d_{Ce}/d_{Ru} in the ground layer of the atmosphere are not clear. Perhaps, it is a particular result. For example, according to data (SHLEIEN, 1965) in the U.S.A. during the period from 9.III.1964 through 20.IX.1964 Ru^{106} and Ce^{144} were distributed equally with aerosol fractions.

Isotopic fractionation with particle size and problems of global transport of aerosols from nuclear tests in the atmosphere

Radioactive aerosol size distribution variations with time and their dependence on meteorological situation in a separate local region and also phenomena of the second isotopic fractionation in the ground layer of the atmosphere were considered above. Observation of late years show that essential peculiarities are observed in the behaviour of separate size fractions of radioactive aerosols and in their transport in the atmosphere in the global scale. It is known that radioactive isotopes forming during nuclear explosions are distributed not equal enough according to aerosol size spectrum. In particular, the larger particle fractions are rich in some isotopes ($Zr^{95} + Nb^{95}$, Ce^{144} W^{185}); finer particle fractions, on the contrary, are rich in others (Sr^{90}, Cs^{137}, Ru^{106} and others) (EDVARSON *et al.*, 1959). That is why transport of one isotopic group in the atmosphere especially in its upper part (stratosphere, mesosphere) may, in principle, differ from that of another group because of different sedimentation rate of their carriers-aerosol particles. Really, in a number of papers some cases were noted when the concentration maximum of gaseous C^{14} in the stratosphere was lower than that of fission products, the maximum of W^{85} was lower than that of Sr^{90} (MACHTA *et al.*, 1959; KAROL *et al.*, 1965). In 1958 the majority of nuclear bomb explosions took place at a relatively not high altitude. Local fall-out (large particles) enrich with Ce^{144} were comparatively large. Therefore fission products depleted with Ce^{144} in comparison to Sr^{90} were released into the stratosphere. On the other hand, in high altitude explosions in 1958 from which there was no local fall-out, fission products injected into the higher layer of the atmosphere had their usual composition. It allowed to notice the appearance of aerosols from nuclear explosions at high altitudes in the upper stratosphere after their descent from the mesosphere and to observe these aerosol transports from the stratosphere into the troposphere up to the ground surface by considering the boundary movement of the area with increased values of concentration ratio Ce^{144}/Sr^{90} (TELEGADAS *et al.*, 1964). High concentration values of Ce^{144} relative to Sr^{90} in radioactive aerosols at high altitudes in the stratosphere can be explained, at least partially, by the fact that the aerosol-carrier of Ce^{144} is

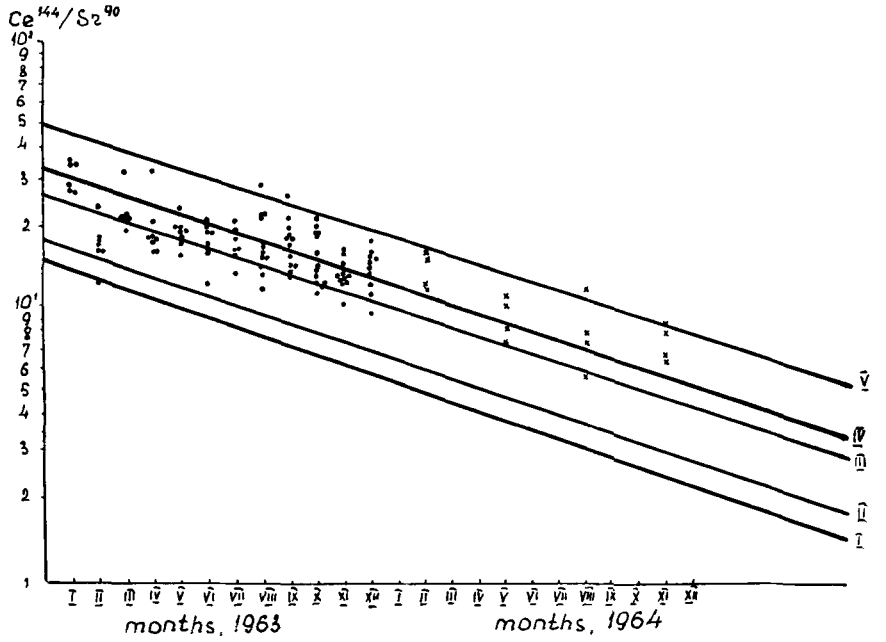

FIG. 7. Ce^{144}/Sr^{90} in the ground air along 80° W.Z. for the northern hemisphere (HASL 155, 1965).

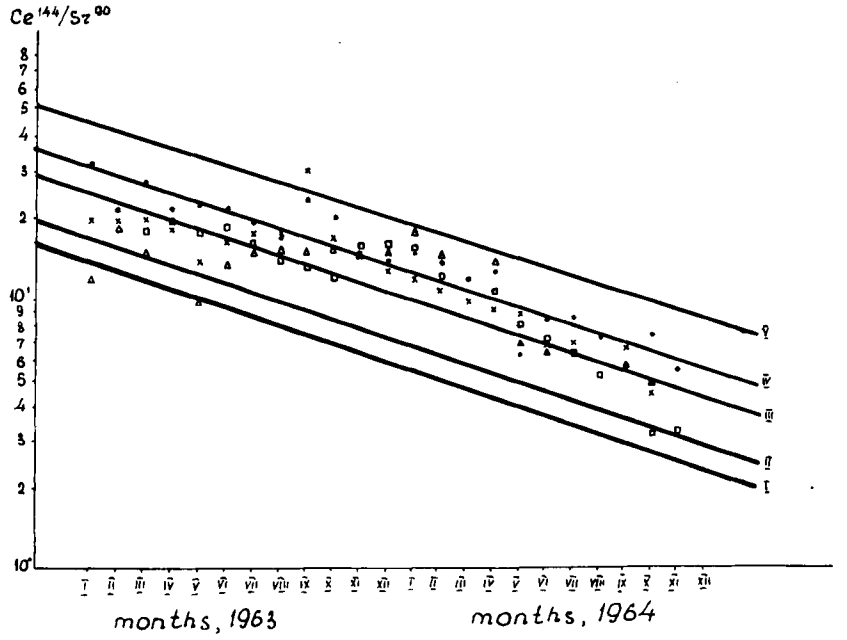

FIG. 8. Ce^{144}/Sr^{90} in the stratosphere in 1963–64. ● 65–70 10^3 ft; × 79–82 10^3 ft; △ 83–91 10^3 ft; □ 100–110 10^3 ft (31° N). I, explosion of 1.09.61; II, explosion of 1.11.61; III, explosion of 1.06.62. IV, explosion of 1.08.62; V, explosion of 1.01.63. (HASL 149, 1964; HASL 158, 1965).

larger than that of Sr^{90} or Cs^{137} and therefore it can descend quicker from the upper layers into the stratosphere. In particular, according to our measurements in the stratosphere in 1961 at the levels of 19–26 km, there was a lack of Cs^{137} relative to Ce^{144} and Sr^{90} (GASIEV *et al.*, 1965*b*). The Cs^{137}/Sr^{90} ratio was on the average close to 1.0. The aerosol-carrier of Cs^{137} according to the existing notions of the mechanism of radioactive particle formation during explosions has smaller sizes than that of Ce^{144} and Sr^{90} and that is why by 1961 it could descend (from the mesosphere into the stratosphere) in less quantities than that of Ce^{144} or Sr^{90}.

During series of nuclear explosions in 1961–1962 there was a combination of relatively low-yield explosions at relatively small altitude and of super high-yield explosions during which fission products were thrown into high altitudes (up to 45–50 km) (HASL 142, 1964). We have already mentioned (MALAKHOV *et al.*) that in the fourth quarter of 1963 analogous to the summer of 1961, increased values of Ce^{144}/Sr^{90} ratio in fall-out were observed. This was confirmed by the data of measurements in the air along the 80° W.Z. (Fig. 7) and in the stratosphere of the northern hemisphere between 19 and 27 km (Fig. 8). In the southern hemisphere the increase of the Ce^{144}/Sr^{90} ratio in the stratosphere took place from July through November in 1963 (Fig. 9). In the northern hemisphere the increase of the Ce^{144}/Sr^{90} ratio in autumn, 1963, was accompanied by the concentration increase of Cs^{137}, Sr^{90}, Mn^{54}, Fe^{59} and other isotopes at all levels in the layer from 19.5 to 27 km. As an example the data on concentration changes of some isotopes in the stratosphere at an altitude of 24–25 km in 1963 are given in Fig. 10. On the basis of the presented facts it is possible to conclude that in 1963 the increase of the Ce^{144}/Sr^{90} ratio is also connected with the flux of nuclear explosion products into the lower stratosphere from the upper stratosphere where these products were thrown.

Summary

From the above data the following conclusions can be drawn:

1. If in the atmosphere there are aerosols from nuclear explosions for a rather long time

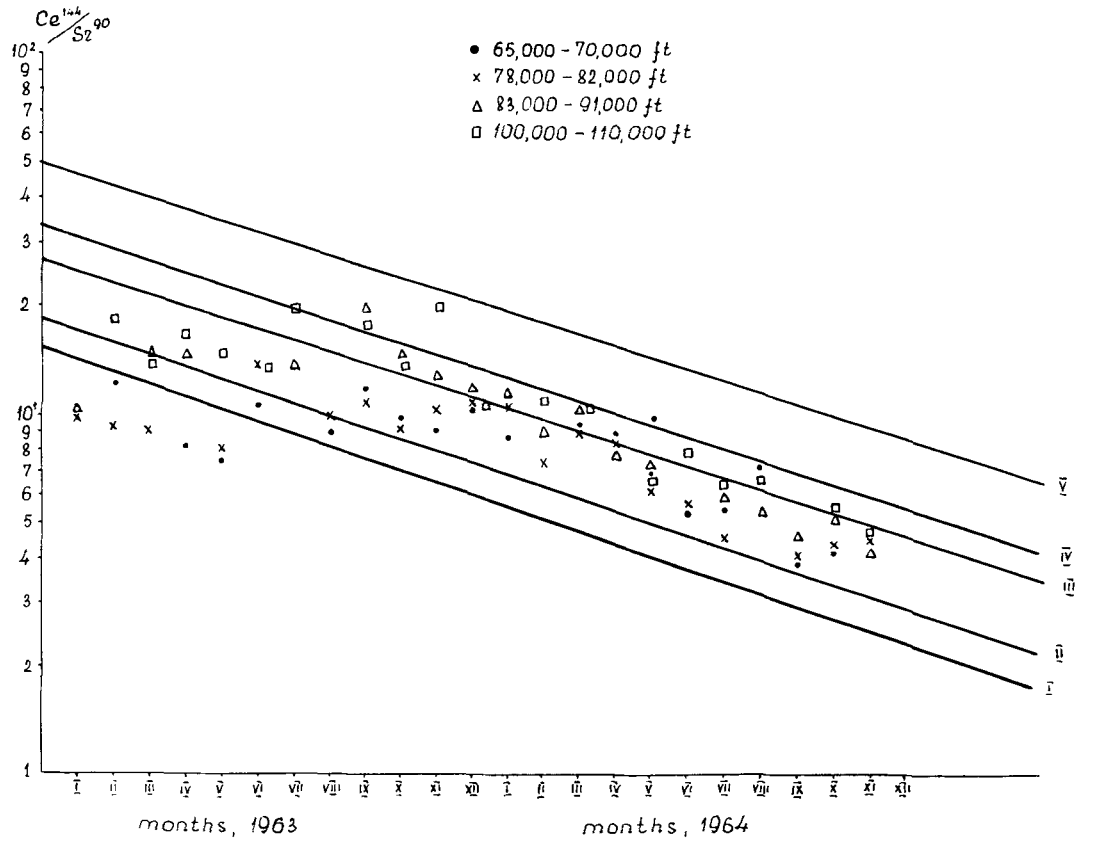

FIG. 9. Ce^{144}/Sr^{90} in the stratosphere during 1963–1964 for the southern hemisphere (32° N). (HASL 149, 1964; 158, 1965.) ● 65–70 10^3 ft; × 79–82 10^3 ft; △ 83–91 10^3 ft; □ 100–110 10^3 ft (31° N).

their size distribution is significantly transformed. Four types of particle size distribution variations were observed.

(*a*) Gradual variation of particle size distribution with time. For example, gradual decrease of the finest particle fraction owing to particle coagulation of this fraction with the particles of natural aerosols.

(*b*) Seasonal variations of particle size distribution caused by fluctuations of air mass exchange between the stratosphere and the troposphere.

(*c*) Fluctuating variations. These are due to simultaneous action of the larger number of different momentary factors: washout, coagulation, small scale horizontal and vertical exchange of air masses in the atmosphere.

(*d*) Impulsive variation of particle size distribution which takes place under rapid entrance of air masses from upper layers of the atmosphere into lower ones.

2. Particle size distribution changed with altitude. Experimental data showed that in December, 1964, the equal amount of radioactivity (about 50%) of aerosols fell on the particles with $d < 0.3$ μ and $d > 0.3$ μ in the middle troposphere and on the particles with $d < 0.5$ μ and $d > 0.5$ μ in the ground layer of the atmosphere. As there is greater difference in mean sizes of radioactive particles for stratospheric and tropospheric aerosols (hundredth and tenth of micron, respectively) the dependence of aerosol particle size distribution on altitude can be used for studying vertical exchange in the atmosphere. It can be especially useful for studying the cases when there is fast flux of air masses from the upper layers of the atmosphere into the lower ones.

3. Experimental data, for example, variation of the Ce^{144}/Sr^{90} ratio, reveal that radioactive aerosol size distribution exerts an essential influence on transport of these aerosols in the atmosphere, especially in the upper layers of the stratosphere and mesosphere.

Results obtained by the present time allow us to hope that in spite of the complexity of connections between particle size distribution and aerosol particle transport in the atmosphere, these connections will be seen more clearly with further accumulation of experimental data. This will allow to study deeper pecularities of transport of different size particles in the atmosphere.

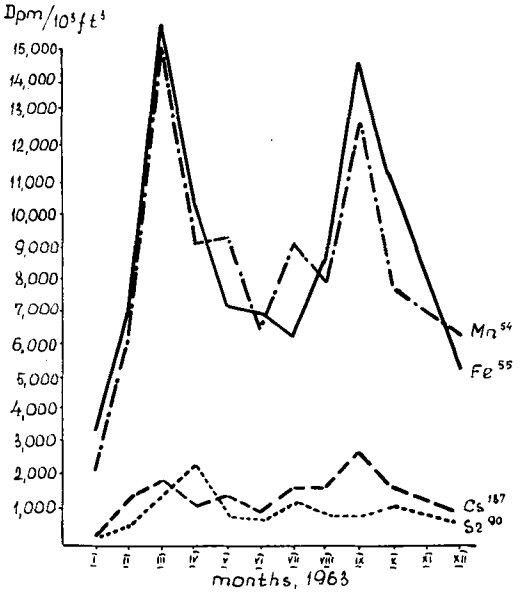

Fig. 10. Mn^{54}, Fe^{55}, Cs^{137}, Sr^{90} concentration at an altitude of 24–25 km during 1963 in San Angelo (31° N). (HASL 149, 1964; 158, 1965.)

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РАСПРЕДЕЛЕНИЕ ПО РАЗМЕРАМ РАДИОАКТИВНЫХ ЧАСТИЦ ОБРАЗОВАВШИХСЯ ПРИ ИСПЫТАНИЯХ ЯДЕРНОГО ОРУЖИЯ И ИХ РАСПРОСТРАНЕНИЯ В АТМОСФЕРЕ

В 1964–65 гг. в Московской области было изучено распределение радиоактивных частиц по размерам в приземном слое атмосферы, а в декабре 1964 года были также проведены измерения на высоте 4–6 км. Наблюдалось 4 вида отклонений распределения радиоактивных аэрозолей по размерам. В тропосфере на высоте 5–6 км радиоактивные частицы были в среднем меньшего размера, чем в приземном слое. На высоте 5–6 км средний диаметр частиц, содержащих Cs^{13} был больше, чем диаметр частиц содержащих Ce^{144}.

Это различие сглаживалось в пробах взятых у поверхности. Был проведен анализ литературных данных о вариациях отношения концентрации различных изотопов. Это показало, что распределение радиоактивных частиц по размерам существенно влияет на их распространение в атмосфере, особенно в высоких слоях стратосферы и мезосферы.