Observations on the particle properties of nuclear debris in the upper atmosphere

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(Manuscript received October 4, 1965)

ABSTRACT

By means of an autoradiographic method, the number concentration of nuclear debris particles of different radioactivity content has been studied. The material consisted of air-filter samples taken at an altitude of 8–12 km in 1961–1963. In the periods studied the concentration of particles with diameters larger than about 0.5 μm was of the order 10–100 kg^{-1} in the lower stratosphere and decreased slowly with time. It was found that the content of such particles relative to the total radioactivity decreases fairly rapidly. It was also observed that the relative concentration of larger particles is higher in the tropospheric samples than in the stratospheric.

Introduction

When nuclear explosions occur in the atmosphere at so high altitudes that no appreciable amounts of material from the ground level are introduced in the explosion cloud, the debris produced consists mainly of quite small particles. Observations on debris from different tests, presumably of this type, and theoretical considerations indicate that a large part of the radioactivity produced is carried by particles with diameters of a few tenths of a μm and less, and particles larger than some μm are usually quite rare (cf. DREVINSKY & MARTELL, 1962; EDVARSON, 1965; SCHUMANN & BAUST, 1965; SISEFSKY, 1961; and STEWART, 1956). Quantitative information about the particle properties of debris of this type is rather scarce, especially as regards the number concentration of particles in the atmosphere. In this paper is presented a study of the concentration of radioactive particles in the atmosphere during two half-year periods following test series.

Material

During a test period, the concentration and size spectrum of radioactive particles is often highly variable, due to injection of new debris with irregular intervals, but quite soon after the end of such a period a more regular pattern is observed. For this reason a number of upper-air samples were chosen from the periods 1.12.1961–23.7.1962 and 1.1–30.9.1963, both following USSR tests in the Arctic with considerable injections of radioactive material (HARDY *et al.*, 1964). The samples were taken with a jet-plane carried device, using glass-fibre filter, described in (EDVARSON *et al.*, 1960). As a rule, two samples were taken within a time of two hours, one 1 km above and one 1 km below the tropopause altitude, as reported by the weather service. The samples have been classed accordingly as stratospheric and tropospheric. As the tropopause is quite often ill defined, and further, changes in the weather situation sometimes have caused that the actual tropopause altitude probably deviates considerably from the reported, this rather rough method has certainly led to a wrong classification in a number of cases.

Methods of measurement

The upper-air samples are analyzed on a routine basis by gamma-spectrometric and autoradiographic methods which have been described in other reports (EDVARSON *et al.*, 1960; EDVARSON & LÖW, 1965; SISEFSKY, 1964). For the present study it was decided to make a special autoradiographic exposure,

TABLE 1. *Data regarding the autoradiographic procedure.*
X-ray film: Ilford Ilfex. Developer: Phen-X.

Sampling period	Autoradiography	Code	Class limits (p Ci)	Approximate particle diameter (μm)
			$6.9 \cdot 10^{-3}$[a]	0.2
1.12.61–24.4.62	June–Juli 62 (42 d)	A 1	$55 \cdot 10^{-3}$	0.4
		A 2	$440 \cdot 10^{-3}$	0.7
8.5.62–23.7.62	Nov.–Dec. 62 (44 d)	A 3	3.55	1.5
		A 4	27.3	2.8
			$4.9 \cdot 20^{-3}$[b]	0.2
3.1.63–30.9.63	Nov. 63–Jan. 64 (77 d)	B 1[c]	$31.5 \cdot 10^{-3}$	0.4
		B 2[c]	$180 \cdot 10^{-3}$	0.6
		B 3	1.90	1
		B 4	12.3	2
		B 5		

[a] In June 1962.
[b] In November 1963.
[c] No data given in this report.

where all samples from one period were exposed at the same time. In this way these autoradiograms were directly comparable. The first series was, however, divided in two parts, which were autoradiographed at different times. In this case a number of samples were exposed both times and their autoradiograms used for intercalibration. The sizes of the spots formed on the autoradiograms were then used to estimate the radioactivity of the individual particles according to a method reported earlier by one of us (SISEFSKY, 1961), and the particles were sorted in a number of radioactivity classes. It was found that the most reproducible way to do this, was to use standard spots, representing the class limits, and compare these spots with the spots to be classed, directly under the microscope. The details about the exposures and radioactivity classes are given in Table 1.

The filter area autoradiographed was about 200 cm^2, as a rule corresponding to a filtered air mass of 10–20 kg. The number of particles in the higher classes was usually rather low and in these classes all particles were counted. In the lower classes, the total number of particles was often quite high, and then about 200 particles were counted in small areas spaced over the autoradiogram.

As there is a relation between the size of the particles and their content of radioactivity, the radioactivity classes used will very roughly correspond to size classes. The range observed in particle radii for a given content of radioactivity is of the order $\pm 30\%$ and varies for different test. In order to give a qualitative information of the particle sizes the mean particle diameters, corresponding to the class limits used, have been estimated from the radioactivity-size relations observed in fresh debris from the USSR-tests in 1961 and 1962 (SISEFSKY, 1964). These values are also given in Table 1.

It is of interest to relate the number concentrations obtained to the radioactivity concentration of the air. The radioactivity values were obtained from remeasurements of the "total γ-activity" of the parts of the filters that had been used for γ-spectrometric analysis earlier. Most of the samples from one period were remeasured at about the same time in order to make the values comparable without corrections for decay. The "total γ-activity" is a well-defined measure of the content of radioactivity only when the composition of the sample is known, and is thus not suitable for absolute assessments. In the present material it was found from the γ-spectrometric determinations, which will be reported elsewhere, that the composition of the samples within one series only varied within such limits that the "total γ-activity" gives a quite good relative measure of the radioactivity concentration.

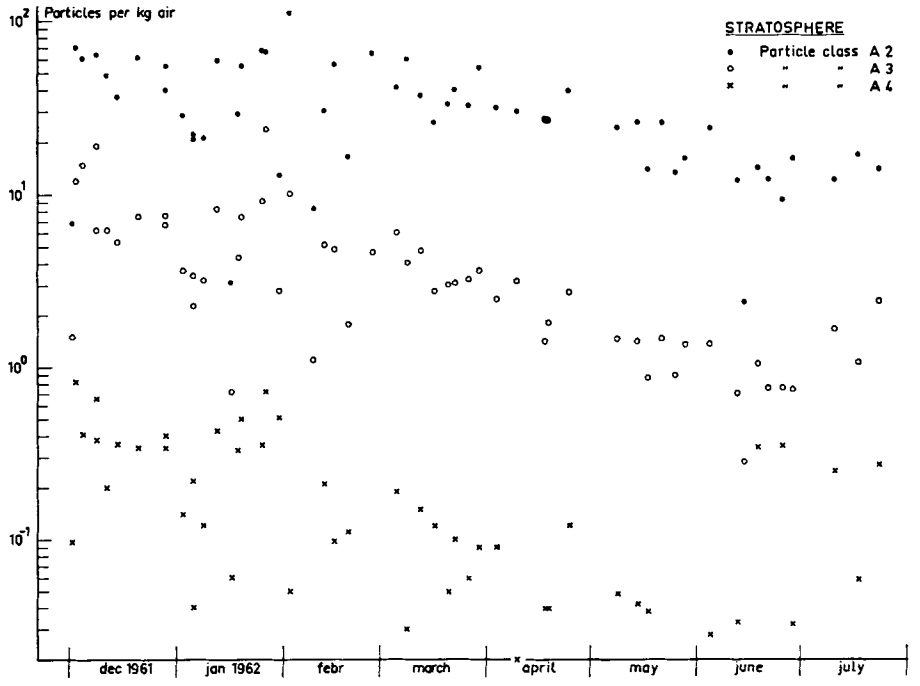

FIG. 1. Particle concentrations in different particle classes.

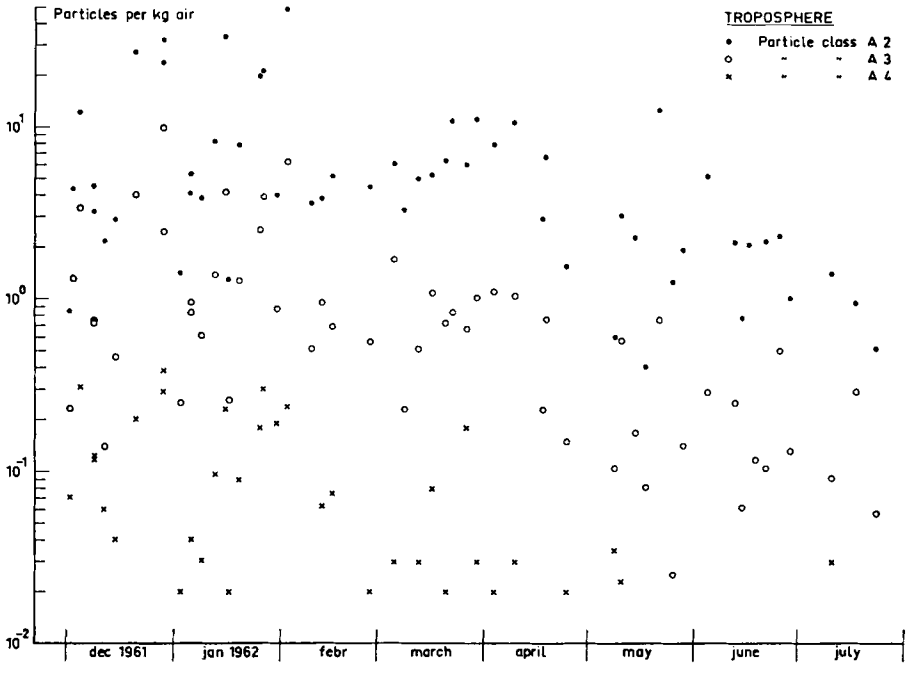

FIG. 2. Particle concentrations in different particle classes.

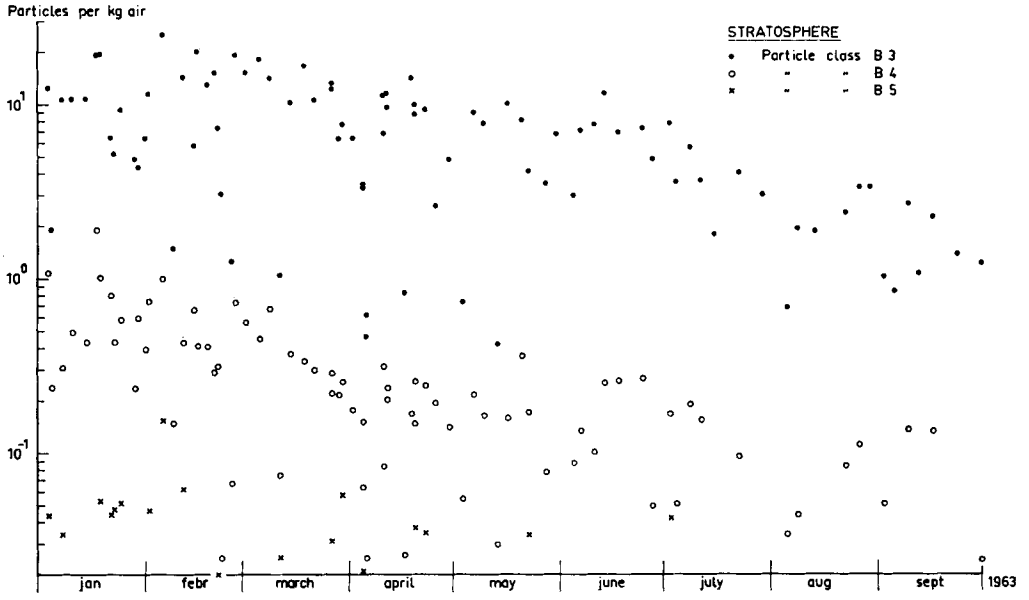

FIG. 3. Particle concentrations in different particle classes.

Results and discussion

The number concentration of particles in the individual samples is given in Figs. 1–4. Table 2 contains monthly mean values, the corresponding coefficients of variation, and correlation coefficients for the paired stratospheric and tropospheric samples. It is seen, that the concentrations decrease slowly with time, and also that the tropospheric values are more scattered than the stratospheric. The number of particles in the highest classes is extremely low; except for the first months of the sampling periods, there are some ten particles or less observed

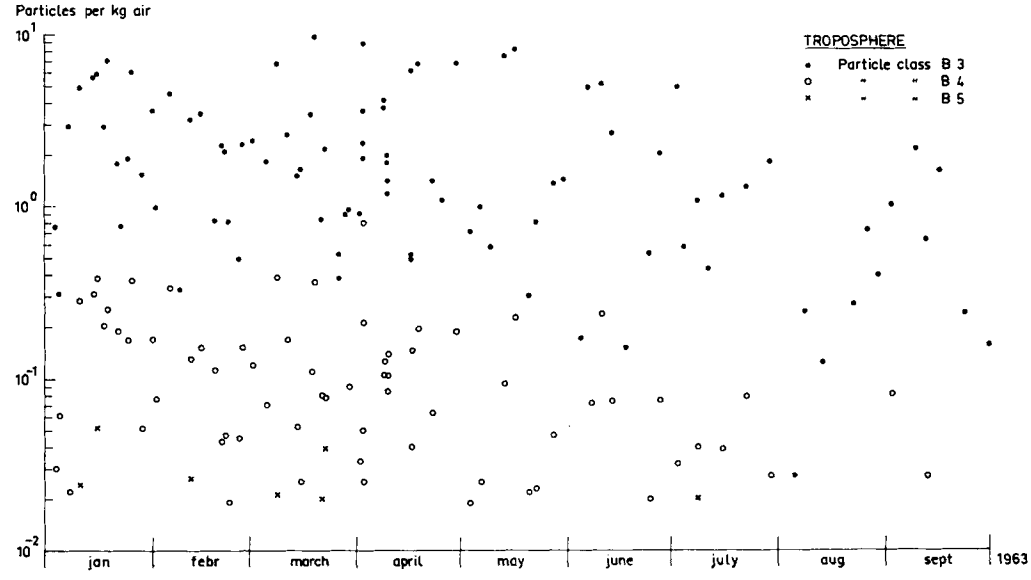

FIG. 4. Particle concentrations in different particle classes.

TABLE 2. *Number of particles per kg air, monthly grouped.*

N: number of samples; *M:* arithmetic mean; *V:* coefficient of variation; *ϱ:* correlation coefficient.

		Stratosphere			Troposphere			Paired samples	
Month	Class	*N*	*M*	*V*	*N*	*M*	*V*	*N*	*ϱ*
Dec. 61	A 1	10	284	0.41	10	51.3	1.12	10	0.18
	2		50.8	0.48		11.3	1.05		0.14
	3		8.64	0.59		2.32	1.28		0.05
	4		0.40	0.52		0.18	0.66		—
Jan. 62	2	11	35.2	0.65	11	10.1	1.02	10	0.84
	3		6.28	1.01		1.54	0.89		0.95
	4		0.31	0.70		0.11	0.91		—
Feb.	2	5	47.9	0.79	6	13.3	1.51	5	0.83
	3		4.62	0.70		1.79	1.39		0.88
	4		0.08	—		0.08	—		—
March	2	8	40.4	0.28	8	6.71	0.41	8	0.10
	3		3.81	0.29		0.84	0.52		0.40
	4		0.10	—		0.05	—		—
April	1	5	448	0.23	5	69.8	0.67	5	0.23
	2		30.8	0.17		5.92	0.62		−0.41
	3		2.32	0.31		0.65	0.68		0.41
	4		0.05	—		0.014	—		—
May	2	6	19.8	0.30	7	3.13	1.35	6	0.50
	3		1.23	0.23		0.26	1.08		0.52
	4		0.02	—		0.008	—		—
June	2	7	12.8	0.51	7	2.20	0.64	7	0.76
	3		0.80	0.41		0.21	0.72		0.30
	4		0.11	—		—	—		—
July	2	3	14.2	—	3	0.95	—	3	—
	3		1.69	—		0.15	—		—
	4		0.19	—		0.029	—		—
Jan. 63	B 3	14	10.1	0.52	13	2.63	0.81	13	0.60
	4		0.64	0.69		0.110	1.00		0.27
	5		0.020	—		0.002	—		—
Feb.	3	12	11.4	0.68	11	1.92	0.71	11	0.68
	4		0.44	0.68		0.101	0.92		0.85
	5		0.024	—		0.002	—		—
March	3	11	11.5	0.44	13	1.99	0.85	11	0.24
	4		0.34	0.50		0.090	1.13		0.60
	5		0.010	—		0.006	—		—
April	3	16	6.97	0.59	18	2.70	0.88	6	−0.27
	4		0.17	0.51		0.12	1.51		−0.10
	5		0.006	—		—	—		—
May	3	9	6.41	0.47	9	1.63	1.50	9	0.44
	4		0.14	0.75		0.044	1.57		0.08
	5		0.004	—		—	—		—
June	3	7	6.94	0.38	7	2.23	0.96	7	0.37
	4		0.165	0.56		0.068	1.21		−0.38
July	3	7	4.25	0.46	7	1.61	0.96	7	0.74
	4		0.094	—		0.031	—		—
	5		0.006	—		0.003	—		—
Aug.	3	6	2.26	0.45	6	0.30	0.82	6	0.87
	4		0.046	—		—	—		—
Sept.	3	7	1.50	0.47	7	0.83	0.97	7	0.87
	4		0.049	—		0.015	—		—

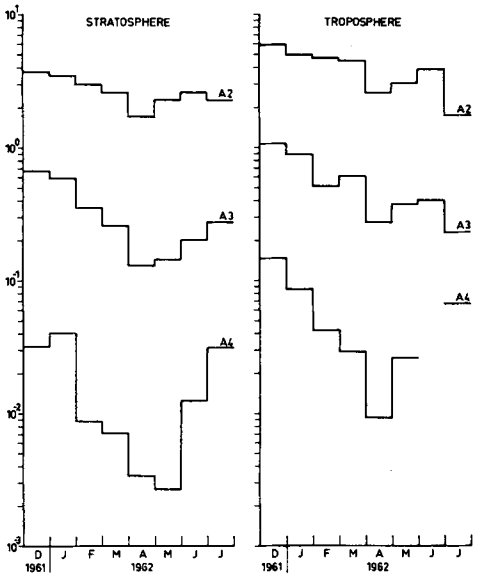

FIG. 5. Monthly averages of the number of particles per unit amount of radioactivity (relative units).

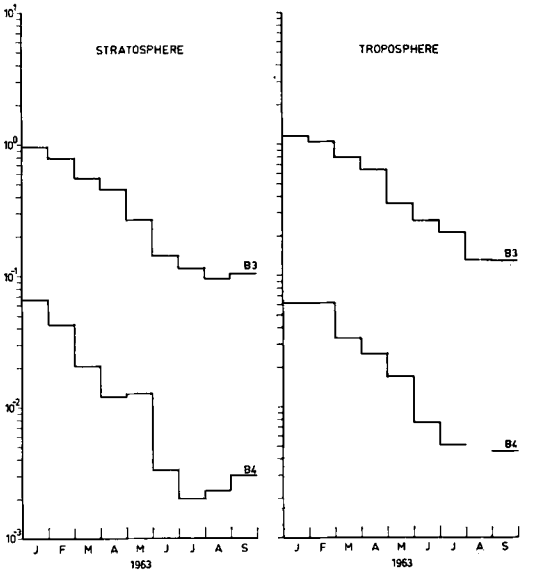

FIG. 6. Monthly averages of the number of particles per unit amount of radioactivity (relative units).

per month in the material, which means that the concentrations given for classes A4 and B5 are only to be regarded as qualitative. The correlation coefficients are as a rule positive. With the small number of samples per month available, these values must be regarded with caution. It is notable, however, that the coefficients are close to zero or negative for the classes A1, A2 and B3 in the spring months March–April for both the periods studied.

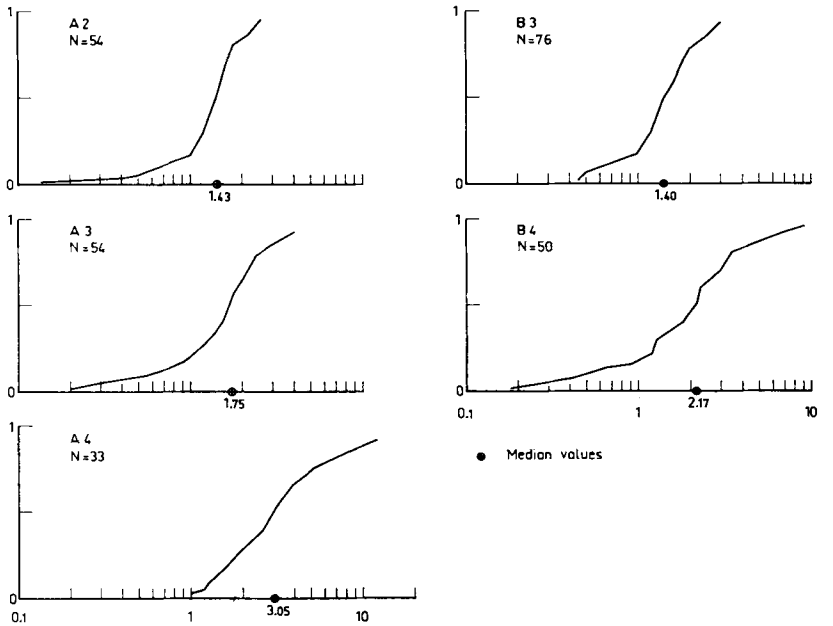

FIG. 7. Cumulative distributions of the ratio between tropospheric and stratospheric number of particles per unit amount of radioactivity.

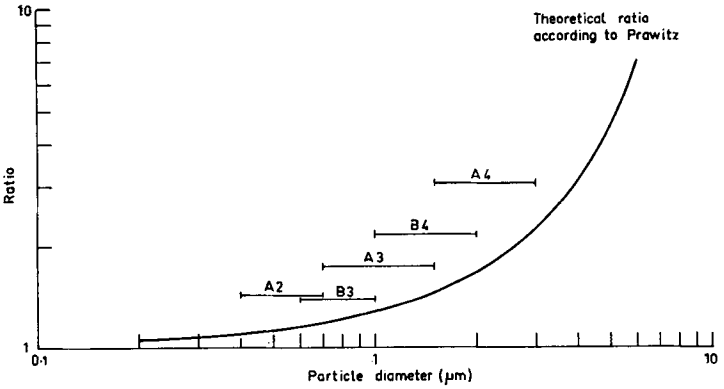

FIG. 8. Comparison between observed concentration ratios and theoretical values according to PRAWITZ (1964).

In Figs. 5 and 6 the number of particles per unit amount of radioactive material has been plotted. These figures show clearly, that the relative amount of particles in the classes studied decreases relatively fast with time. In the samples from 1962 there was an increase in the higher classes in June and July, which might be due to a contribution from the US tests at Christmas Island in the spring of 1962.

In Fig. 7, the ratio between the number of particles per unit amount of radioactivity in the paired stratospheric and tropospheric samples are plotted. It was found that this ratio did not vary in a systematic way with time, and for that reason it seemed permissible to bulk the values for the whole sampling periods.

It is seen that this ratio is centered well above one, and further, that it increases with class number. A possible explanation to this is that the transport process in the tropopause layer will cause a selection with particle size in the way studied theoretically by PRAWITZ (1964). From the calculations reported by Prawitz, theoretical ratios corresponding to the observed values given in Fig. 7 can be obtained if the particle size spectrum is known. For a rough, qualitative comparison the observed median values and the theoretical concentration ratios for different particle sizes divided by the concentration ratio for particles of diameters < 0.2 μm have been plotted in Fig. 8.

Acknowledgements

A considerable part of the spot counting was made at The State Control Institute for Agricultural Chemistry under the supervision of Dr Britt Hagström. The data processing and statistical calculations were made by Miss Kerstin Hallgard at this institute. We want to express our gratitude for the very competent help we had from them.

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ИЗУЧЕНИЕ СВОЙСТВ ПРОДУКТОВ ЯДЕРНОГО РАСПАДА В ВЕРХНЕЙ АТМОСФЕРЕ

С помощью авторадиографического метода была изучена концентрация частиц продуктов ядерного распада, с различной радиоактивностью. Пробы профильтрованного воздуха брались на высоте 8–12 км в течение 1961–1963 годов.

В этот период концентрация частиц диаметром больших, чем 0,5 μm была в нижней стратосфере порядка 10–100 кг$^{-1}$ и уменьшалась со временем. Найдено, что концентрация таких частиц уменьшается довольно быстро по сравнению с полной радиоактивностью. Наблюдалась также, что относительная концентрация крупных частиц в тропосферных пробах более высокая, чем в стратосферных.