

# Properties of the stratospheric aerosol

BY JAMES P. FRIEND, *Isotopes, Inc., Westwood, New Jersey*<sup>1</sup>

(Manuscript received November 9, 1965)

## ABSTRACT

Studies of the chemical composition, size distributions and concentrations of stratospheric aerosol particles have shown that the particles consist of ammonium sulfate and ammonium persulfate, and may have liquid associated with them in the stratosphere. The average particle size distribution function was found to approximate a log-normal distribution over the range of radius 0.1 to 1.0 microns. The number concentrations found were variable but generally of the order of  $0.05 \text{ cm}^{-3}$  at 18 km altitude. The corresponding mass concentrations are of the order of  $10^{-2} \text{ pg cm}^{-3}$ . Combination of the presently determined size distribution with estimates of the size distribution of Aitken nuclei in the stratosphere indicates that the overall distribution of particulate matter in the lower stratosphere may be bimodal. This bimodality indicates that other processes in addition to coagulation must occur if the particles with radius  $< 0.1$  micron give rise to particles with radius  $> 0.1$  micron.

## 1. Introduction

Aircraft sampling of stratospheric aerosols was first undertaken in 1960 and the results were presented by JUNGE & MANSON (1961) and FRIEND *et al.* (1961). JUNGE, CHAGNON & MANSON (1961) had performed balloon sampling of stratospheric particles, and had found that the preponderance of particles in the 15 to 25 km altitude region contained sulfur, and had radii in the region 0.1 to 1.0 micron. FRIEND *et al.* (1961) found that the particles consisted primarily of ammonium sulfate and possibly ammonium persulfate. JUNGE, CHAGNON & MANSON (1961), JUNGE & MANSON (1961), and FRIEND *et al.* (1961) all concluded that the frequency distribution of radii of the particles was monotonic, the number of particles decreasing with increasing radius from 0.1 micron to 1.0 micron. Few, if any, sulfate particles were observed with radii greater than 1.0 micron.

MOSSOP (1965) has recently reported results of aircraft sampling of particles collected in early 1963. He found that the sulfate particles, which are water soluble, contain one or more particles ranging in radius from 0.005 to 0.4 micron which are insoluble in water. He also found that the frequency distribution of radii was peaked in the vicinity of 0.3–0.4 micron.

The results reported and discussed in this paper concern samples collected between January 1962 and August 1963 by WU-2 aircraft. It

is the purpose of the present paper to discuss what is currently known of the physical properties of the stratospheric aerosol, and what factors are significant with regard to the origin and behavior of the particles.

The work reported here was performed as part of the Project Stardust High Altitude Sampling Program sponsored by the Defense Atomic Support Agency of the U.S. Department of Defense under contract No. DA-49-146-XZ-079.

## 2. Particle sampling and analytical methods

The methods of sampling, analysis and converting observed particle size distributions to those as they exist in the atmosphere have been described in detail elsewhere (FEELY *et al.*, 1963). For the purpose of this paper, it is sufficient to note the following items:

(a) The particles were collected at the desired altitude using an impaction probe which permitted exposing prepared impaction surfaces of carbon on nitrocellulose film to the free air stream.

(b) The characteristic impaction dimension, i.e. the width of the impaction surface, was 0.3 cm.

(c) After sampling for the desired length of time, the impaction surfaces were kept protec-

TABLE 1. *Composition of stratospheric particles in impaction samples.*

Sample No.	Sampling date	Altitude (km)	Latitude	Composition <sup>a</sup>
241	25 Jan. 62	21	30° N	S
242	30 Jan. 62	18	31°-48° N	S
243	13 Feb. 62	18	31°-48° N	S
244	13 Feb. 62	15	31°-48° N	P
245	13 Mar. 62	12	31°-48° N	P
251	5 June 62	18	31°-48° N	P
252	19 June 62	15	31°-48° N	S
254	12 June 62	17	31°-48° N	S
257	26 June 62	17	31°-48° N	M
258	14 Feb. 63	18	39°-45° N	S
259	19 Feb. 63	18	31°-19° N	S
260	6 Feb. 63	20	32°-56° N	P
262	21 Feb. 63	20	49°-64° N	S

<sup>a</sup> S = Ammonium sulfate; P = Ammonium persulfate; M = Mixture of S and P.

ted in the air-tight body of the probe until opened in the laboratory.

(d) The particles were observed in an electron microscope and electron diffraction patterns of single particles or a few particles in a small area of the sample were obtained.

(e) The sulfate particles (which comprise almost all of the mass of the stratospheric aerosol) were, to a large degree, flat rosettes, and often were accompanied by haloes of very small particles.

(f) It was assumed that the particles were spherical prior to collection and the observed sizes were then corrected for flattening using

the further assumption that the particles with observed radius greater than 0.2 micron had a thickness of 0.2 micron as had been indicated in electron micrographs of shadowed deposits.

(g) These calculations were used to convert the observed number of particles in a given size class on a given area of sample to concentrations. By using flight data, the width of the impaction surface, and applying the RANZ & WONG (1952) impaction theory for an ideal ribbon, the collection efficiency for each size class of particle was determined. The particle density used was 2 g/cm<sup>3</sup>.

### 3. Composition and concentrations

Table 1 lists the chemical compositions of those samples for which selected area electron diffraction results were obtained.

The particle number concentrations and the corresponding mass concentrations (using a particle density of 2 g/cm<sup>3</sup>) are listed in Table 2 along with the sampling dates and locations.

### 4. Size distribution function

The size-frequency spectrum, or size-frequency distribution is often presented in plots of  $d\bar{n}/d(\log r)$  vs.  $r$ , where  $\bar{n}$  is the number concentration of particles and  $r$  is radius of particles. The dimension of  $d\bar{n}/d(\log r)$  is cm<sup>-3</sup>, and the area under a curve (spectrum) is the number concentration. The size distribution function, as defined here, is dimensionless and is essentially the size-frequency distribution

TABLE 2. *Particle number and mass concentrations.*

Sample No.	Collection date	Altitude (km)	Latitude	Longitude	Number concentration (cm <sup>-3</sup> )	Mass concentration (pg/cm <sup>3</sup> )
243	13 Feb. 62	18	33° N	100°-112° W	.045	$6.8 \times 10^{-3}$
258	14 Feb. 63	18	40° N	104°-109° W	.069	$1.7 \times 10^{-2}$
259	19 Feb. 63	18	31°-19° N	100°- 96° W	.059	$1.0 \times 10^{-2}$
265	2 Apr. 63	18	31°-32° N	99°-100° W	.035	$1.2 \times 10^{-2}$
266	7 May 63	18	31°-33° N	100° W	.022	$1.0 \times 10^{-2}$
268	30 July 63	18	32°-33° N	111° W	.023	$5.0 \times 10^{-2}$
271	13 Aug. 63	18	32° N	111° W	.051	$1.3 \times 10^{-2}$
254	12 June 62	17	31°-49° N	101°-112° W	.022	$5.7 \times 10^{-3}$
257	26 June 62	17	31°-48° N	101°-112° W	.020	$4.8 \times 10^{-3}$
244	13 Feb. 62	15	32°-49° N	100°-112° W	.012	$2.7 \times 10^{-3}$
252	19 June 62	15	31°-49° N	101°-112° W	.012	$2.6 \times 10^{-3}$
245	13 Mar. 62	12	49°-31° N	112°- 10° W	.014	$3.3 \times 10^{-3}$

TABLE 3. *Experimental and average values of the size distribution function  $(1/\bar{n})[d\bar{n}/d(\log r)]$ .*

Sample	Sampling date	Alt. (km)	Mean (logarithmic) radius of size class ( $M$ ):										
			.138	.160	.182	.199	.224	.244	.264	.282	.300	.318	.336
243	13 Feb. 62	18	0.356	1.10	1.82	1.58	2.53	4.49	4.52	5.19	2.26	2.19	1.02
258	14 Feb. 63	18	0.041	0.262	0.328	0.397	1.06	1.88	5.57	5.87	4.60	4.36	3.00
259	19 Feb. 63	18	0.519	1.28	1.05	0.857	3.20	4.75	4.11	4.83	1.84	2.44	1.20
265	2 Apr. 63	18	0	0	0	0.488	0.169	0.737	1.47	4.43	5.26	2.82	6.89
266	7 May 63	18	0	0	0	0.026	0.201	0.513	1.08	2.73	4.33	4.15	4.88
268	30 July 63	18	0	0.202	2.39	4.79	3.14	2.76	1.80	1.62	1.69	1.93	1.42
271	13 Aug. 63	18	0	0.114	1.09	0.561	1.89	1.78	3.60	3.40	3.54	4.54	5.00
254	12 June 62	17	0.334	0.618	0.90	0.401	0.498	1.49	2.35	3.38	5.60	6.00	5.06
257	26 June 62	17	0.068	0.361	0.92	0.846	0.652	1.46	4.17	7.01	6.43	3.37	2.61
244	13 Feb. 62	15	0.240	0.72	0.667	0.92	0.992	2.35	3.46	4.74	4.93	5.41	3.98
252	19 June 62	15	1.48	1.72	2.24	2.70	2.13	0.150	0.720	3.19	3.04	2.70	2.31
245	13 Mar. 62	12	0	0	0	1.05	0.664	2.77	2.74	5.98	6.96	5.18	6.41
Median Value			0.055	0.312	0.910	0.863	0.999	1.83	3.10	4.59	4.47	3.76	3.49
Average			0.253	0.531	0.95	1.22	1.43	2.09	2.96	4.36	4.21	3.76	3.65
			.352	.370	.386	.402	.418	.432	.448	.462	.478	.492	.506
243	13 Feb. 62	18	0.978	0.784	0.352	0.392	0.158	0.395	0.030	0.039	0.019	0.013	—
258	14 Feb. 63	18	2.52	1.71	2.33	0.794	1.53	0.842	0.272	0.605	0.157	0.386	0.140
259	19 Feb. 63	18	1.02	0.604	0.916	0.682	0.565	0.528	0.082	0.086	0.058	0.047	0.031
265	2 Apr. 63	18	5.66	4.73	4.74	3.62	0.915	0.510	0.112	0.259	0.089	0.317	0.160
266	7 May 63	18	3.53	3.64	3.19	3.98	3.20	2.92	1.97	2.07	2.35	1.15	1.09
268	30 July 63	18	1.90	1.70	1.38	1.37	0.891	0.861	0.638	0.422	0.307	0.608	0.342
271	13 Aug. 63	18	2.97	3.32	1.68	0.570	0.642	1.95	1.99	0.343	0.351	0	—
254	12 June 62	17	2.90	3.60	1.92	1.78	0.487	0.952	0.490	0	0.023	0	0.287
257	26 June 62	17	2.85	1.91	1.55	0.827	0.618	0.635	0.242	0.210	0.175	0.235	0.478
244	13 Feb. 62	15	3.54	1.75	1.83	0.044	0.094	0.096	—	—	—	0.058	—
252	19 June 62	15	1.25	0.046	0.802	1.40	0.776	0.095	0.876	0.098	2.37	0.067	0.949
245	13 Mar. 62	12	3.67	1.08	0.415	0.543	0.167	0.047	0.090	0.049	0.136	0	0.055
Median Value			2.88	1.73	1.62	0.811	0.630	0.582	0.272	0.210	0.157	0.063	0.224
Average			2.73	2.07	1.76	1.33	0.837	0.819	0.566	0.348	0.504	0.240	0.394
			.520	.534	.548	.560	.574	.588	.600	.614	.626	.640	.652
243	13 Feb. 62	18	—	—	—	—	—	—	—	—	—	—	—
258	14 Feb. 63	18	0.072	0.074	—	—	—	—	—	—	—	—	—
259	19 Feb. 63	18	0.067	0.017	0.055	0.018	0.017	0.019	0.019	0.040	0.020	0.020	—
265	2 Apr. 63	18	0.233	0.439	0.038	0.038	0.147	0.398	0.405	—	—	—	—
266	7 May 63	18	1.05	0.764	0.922	0.257	0.888	0.352	0	0.091	0.373	0.094	0
268	30 July 63	18	0.140	0.144	0	0.080	0.303	0.331	0.084	0.086	0.088	0	0.096
271	13 Aug. 63	18	—	—	—	—	—	—	—	—	—	—	—
254	12 June 62	17	—	—	—	—	—	—	—	—	—	—	—
257	26 June 62	17	—	—	—	—	—	—	—	—	—	—	—
244	13 Feb. 62	15	—	—	—	—	—	—	—	0.073	—	—	—
252	19 June 62	15	0.888	0.072	0.991	0.040	0	0	0	0.043	—	—	—
245	13 Mar. 62	12	0.032	—	—	—	—	—	—	—	—	—	—
Median Value			0.140	—	—	—	—	—	—	—	—	—	—
Average			0.207	0.126	0.167	0.0361	0.114	0.0917	0.0423	0.0278	0.0401	0.010	—

normalized by dividing by the number concentrations, namely:

$$\frac{1}{\bar{n}} \frac{d\bar{n}}{d(\log r)}.$$

In practice, in making and reporting particle size measurements on samples, the number of particles,  $n_i$  in a specified size class, say  $r_a < r < r_b$ , is recorded. The class intervals in this work were chosen such that  $\Delta(\log r)$  is constant.

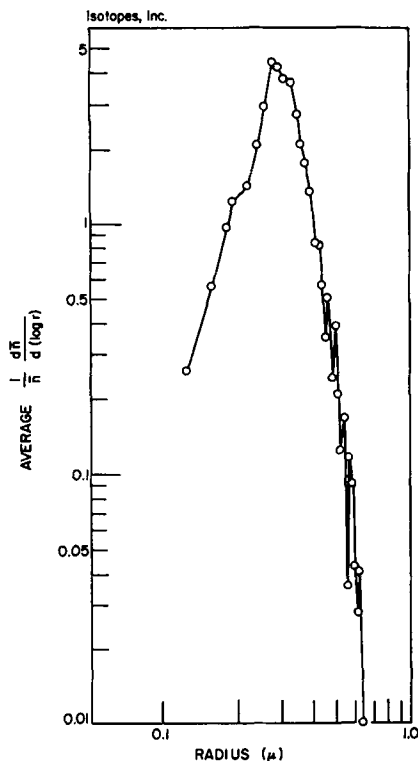


FIG. 1. Average size distribution for the stratospheric aerosol.

When  $n_i$  was converted to number concentration,  $\bar{n}_i$ , by means of flight data and corrections for impaction efficiency, the quantity  $\bar{n}_i/\Delta$

( $\log r$ ) resulted. This is the experimental approximation of  $d\bar{n}/d(\log r)$ . The number concentration for each sample, as reported in Table 2, was obtained by:

$$\bar{n} = \sum \left[ \frac{\bar{n}_i}{\Delta(\log r)} \right] \Delta(\log r).$$

Table 3 lists for each sample the values of the size distribution function for each size class. The size classes are listed according to their mean (logarithmic) radius, in microns. The listed quantity is actually  $(1/\bar{n})[\bar{n}_i/\Delta(\log r)]$ . Listed in the last two rows of the table are the median and average values, respectively, of the size distribution function. The significance of the differences between these two rows will be discussed below. The average distribution function is shown plotted in Fig. 1. The nearly parabolic shape of this function suggests a log-normal distribution. The average distribution function is shown as points in Fig. 2 on a log-probability plot. The straight line drawn among the points is a log-normal distribution with geometric mean radius 0.305 micron and a geometric standard deviation of 1.30.

## 5. Evaluation of measurements

Uncertainties in the data as they are given in Tables 2 and 3 stem from (a) possible non-representativeness of sample areas studied; (b) improper correction for effects of moisture (flattening) on the particles; (c) application of impaction theory for an ideal thin ribbon to the

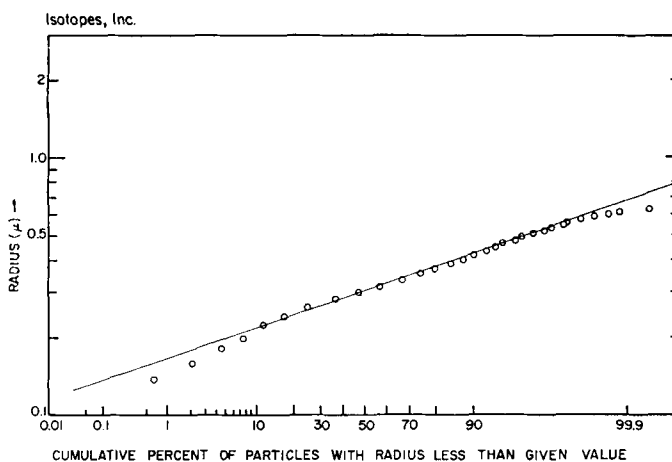


FIG. 2. Average distribution function of stratospheric sulfate particles.

computation of probe collection efficiencies; (d) statistical fluctuations because of small numbers of particles observed in particular class intervals.

The net uncertainty due to the four sources listed above is difficult to assess quantitatively. However, items *a*, *b* and *c* probably cause the distribution functions to be more uncertain for the smaller than for the larger particles. To some degree all uncertainties are reflected in the variations of the experimental values of  $(1/\bar{n})[d\bar{n}/d(\log r)]$  in each column of Table 3. These latter variations in turn are reflected in differences between the median and average values of the distribution function. It must be noted, however, that variations in the values listed in Table 3 may be due to natural variations in production and removal rates of the particles. The relative proportions of real and experimental variations cannot be assessed.

The best established fact of the size distribution function of the stratospheric sulfate particles is the peak in the vicinity of 0.3 micron radius. It is notable that in the range of radius 0.18–0.40 micron:

- the value of the average distribution function increases and then decreases by a factor of four;
- the difference between the average and mean distribution functions varies by less than 30 % of the average;
- the computed values of the collection efficiencies (RANZ & WONG, 1952) at 18 km altitude vary by a factor of about 1.3.;
- the number of observed particles in each size class was greater than in any class outside the size range (0.18–0.40  $\mu$ ).

Thus all of the factors which produce uncertainty in the values of the size distribution function are minimal over the size range in which the peak is centered.

## 6. Comparison with other findings

JUNGE *et al.* (1951) found that the number concentrations of particles with  $0.1 < r < 1.0$  micron had a broad maximum at about 20 km altitude. Since the highest altitude covered in the present work was 18 km, the occurrence of peak concentrations at 20 km could not be demonstrated. However, the decrease of concentration with decreasing altitude from 18,

as shown in Table 2, is in general agreement with Junge *et al.*

More recent work by NEWKIRK & EDDY (1964), using light scattering measurements by a balloon-borne coronagraph, showed that the stratospheric aerosol can exist in thin cloud-like laminae. (Similar conclusions on the existence of layers of aerosols in the stratosphere were drawn by several other workers. These are reviewed by Newkirk & Eddy.) Again, the present work was not extensive enough in time or altitude coverage to demonstrate the existence of such aerosol clouds. The two high values in number concentrations (Samples 266 and 268) were obtained after the eruption of Mt. Agung in Bali, which may have contributed to the number of particles or enhanced particle formation in the stratospheric air in which the samples were taken. (See also MOSSOP (1964) for discussion of the association of sulfate with the volcanic dust). However, the variations by nearly an order of magnitude in number concentrations of the samples from 18 km altitude, as shown in Table 2, are consistent with the concept of clouds of aerosols. It would be very difficult to attribute all observed variations to measurement errors. Since the sampling was not sufficiently intensive, the number concentrations in Table 2 cannot be used to give reliable time-averaged values.

In one important aspect the present work is not in agreement with previous work of JUNGE *et al.* (1961) and JUNGE & MANSON (1961). This is in the form of the size distribution function of the stratospheric sulfate particles ( $0.1 < r < 1.0$  micron). JUNGE *et al.* (1961), from examination of particles collected by balloon-borne impactors, concluded that the radii were distributed approximately according to:

$$\frac{dn}{d(\log r)} \propto r^{-2}$$

or equivalently:

$$\frac{dn}{dr} \propto r^{-3}$$

for the range  $0.1 < r < 1.0$  micron. Fig. 3 shows their distribution and the distribution found in the present work. In Fig. 3 the ordinate,  $dn/d(\log r)$ , has units of  $\text{cm}^{-3}$ . Curve B corresponds to the average distribution function of Fig. 1, and the median value of  $\bar{n}$  at 18 km

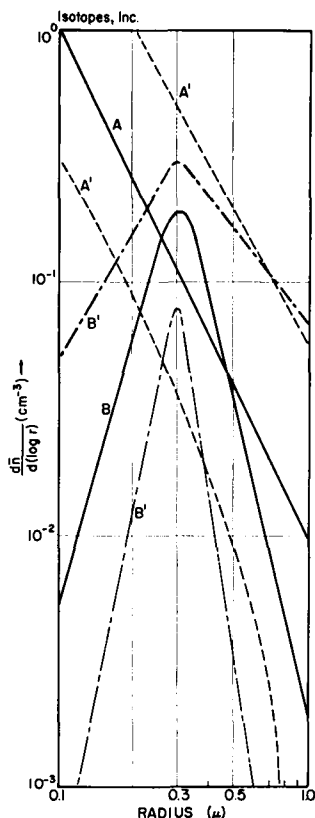


FIG. 3. Comparison of particle size-concentration distributions of stratospheric sulfate particles. Curve A: AFCL—JUNGE *et al.* Curve A': Confidence limits of curve A. Curve B: Present Work. Curve B': Confidence limits of curve B.

altitude, namely  $0.066 \text{ cm}^{-3}$ . Also shown in Fig. 3 are limits of uncertainty estimated by Junge *et al.*, for their curve, and estimated for the present work based on variations as illustrated in Table 3 and discussed above. It can be readily seen that the main region of discrepancy is for  $r < 0.2$  micron.

It is also to be noted that there is a real discrepancy in the shapes of the two distributions shown in Fig. 3. The regions of uncertainty for the present distribution are based primarily on the variations in the absolute values of  $(1/\bar{n}) [d\bar{n}/d(\log r)]$  about the mean value, and do not completely indicate the reliability of the shape of the distribution. The reasons for confidence in the general shape of the size distribution function were discussed at the end of the previous section. It is apparent that the discrep-

ancy between the two distributions in Fig. 3 is due primarily to one or more systematic errors. A likely cause is the correction for the effect of moisture in flattening the particles. JUNGE *et al.* (1961) did not apply any correction for this effect and recorded the sizes of the particles as they appeared in the samples.

MOSSOP (1965) reported the results of analysis of eight WU-2 aircraft impactor samples taken in the latitude range of  $15^\circ$  to  $45^\circ$  South in early 1963. The concentrations ranged from  $0.017$  to  $0.042 \text{ cm}^{-3}$ , and are consistent with all but two of the present results. The size distributions found by Mossop were also peaked, but at a slightly larger radius, about  $0.35$  micron. Fig. 4 shows the present distribution in the same manner as Fig. 3, and also shows Mossop's distribution corresponding to an average concentration of  $0.033 \text{ cm}^{-3}$ . The agreement between the two results is good. The slight difference in the radius at which the peak occurs may be to different methods of correcting for flattening of the particles.

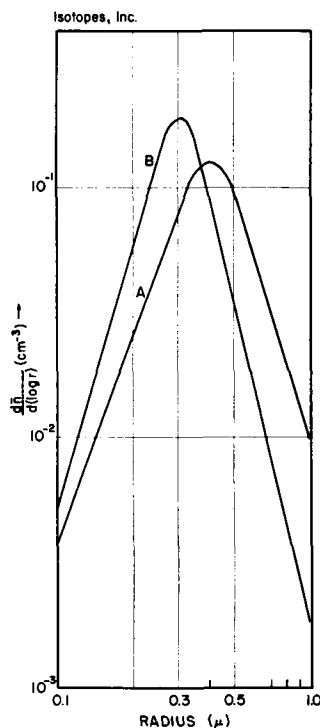


FIG. 4. Comparison of particle size-concentration distributions of stratospheric sulfate particles. Curve A: Mossop, 1965. Curve B: Present Work.

NEWKIRK & EDDY (1964) used measurements of intensity and wavelength of sky light measured by a balloon-borne coronagraph in attempts to deduce the concentrations and size distributions of atmospheric aerosol particles. However, they started by assuming that the functional form of the size distribution was

$$\bar{n} = \bar{n}_0(r/r_2)^{-\delta},$$

where

$$\delta = 0 \text{ for } r \leq r_2, \quad \delta > 0 \text{ for } r > r_2$$

in the manner of JUNGE *et al.* (1961). In (2),  $r_2$  is some fixed radius. The experimental results were then used to calculate values of  $\bar{n}$  and  $\delta$ , which were found to be in substantial agreement with the values reported by JUNGE *et al.* (1961).

In his paper, MOSSOP (1965) suspected that the earlier distributions reported by JUNGE *et al.* and FRIEND *et al.* (1961) might be in error because of the production of small particles in the samples following their collection by condensation of moisture and subsequent drying. Mossop noted that Newkirk & Eddy's apparently independent work did not support this suspicion. So Mossop concluded that the earlier work was also correct and that his collections of particles were made at times and places where the atmosphere was anomalously deficient in particles with  $0.05 < r < 0.4$  micron.

In the light of the present results, and their favorable comparison with Mossop's, the following clarifying statements are in order:

- Mossop's original suspicion was probably correct.
- Since Newkirk & Eddy assumed the wrong functional form of the size distribution, their work does not corroborate the findings of Junge *et al.*
- Mossop's samples were "normal"; i.e. the atmosphere was *not* deficient in particles with  $0.05 < r < 0.4$  micron.

## 7. Aitken nuclei and sulfate particles

Though the present work is not directly concerned with measurement of Aitken nuclei (which consist primarily of particles with  $r < 0.1$  micron in the stratosphere), combining the current results with the known properties of stratospheric Aitken nuclei leads to some interesting conclusions. JUNGE *et al.* (1961) and JUNGE (1961), in experiments with balloon-

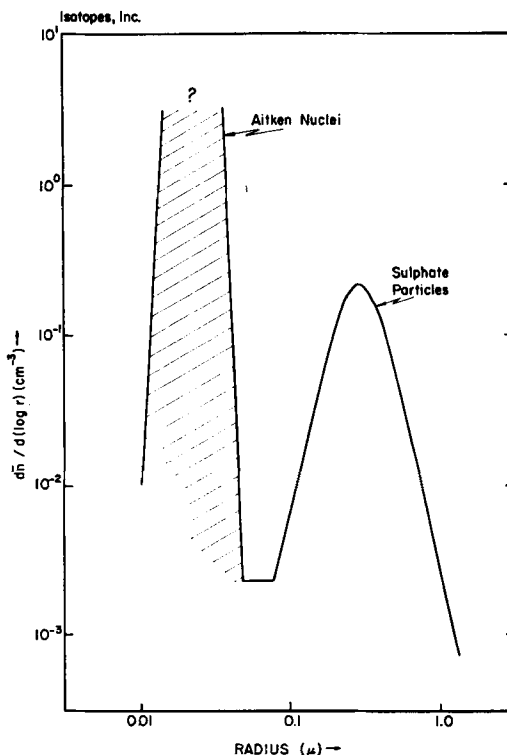


FIG. 5. Schematic size distribution of stratospheric particles including Aitken nuclei.

borne Aitken nuclei counters, found that the total particle concentration decreases rapidly with height above the tropopause for the first few kilometers, and then remains on the order of about  $1 \text{ cm}^{-3}$  at least up to about 33 km altitude. Since Aitken nuclei are smaller than 0.1 micron radius they would not have been collected with appreciable efficiency by the impactors. If the distribution function and number concentrations found in the present work are correct, then in the region of radius less than 0.1 micron the frequency (or number concentration expressed, e.g., as  $d\bar{n}/d(\log r)$ ) must increase again so that the total number concentration will agree with the Aitken nuclei concentration. There is no direct evidence available concerning the average size of the Aitken nuclei. However, JUNGE *et al.* (1961) estimated that a radius of 0.04 micron would be consistent with some theoretical considerations of mixing by turbulent diffusion from a tropospheric source. Thus it is likely that the actual distribution of particle sizes in the strato-

sphere is bimodal with peaks at 0.3 micron and in the vicinity of 0.04 micron radius. Such a distribution is shown schematically in Fig. 5 where the portion of the curve labeled "sulfate particles" corresponds to the present findings and the portion labeled "Aitken nuclei" is conjectural. The integral under the entire curve must be such that the total number concentration is about  $1 \text{ cm}^{-3}$ . The corresponding value for the sulfate particles is  $0.066 \text{ cm}^{-3}$ , as mentioned earlier.

If the general shape of the distribution as shown in Fig. 5 applies to a steady-state condition, then the following statements are true: (a) Some process in addition to coagulation must explain the distribution. (b) If the sulfate particles result from a process involving Aitken nuclei, only some of the Aitken nuclei are involved, and the process is fast compared to coagulation.

## 8. Origin of the sulfate aerosol

The body of data relevant to the stratospheric sulfate aerosol is by no means complete enough to determine mechanisms of its formation and removal. The following discussion shows that the sulfate particles are likely to be created in the stratosphere. For purposes of the discussion the term "large particle" refers to the sulfate aerosol particles.

JUNGE (1965) and CADLE & POWERS (1965) have considered that the photochemical oxidation of  $\text{SO}_2$  can occur in the stratosphere, and that it is the probable means of production of the solid sulfate particles. The finding in this and previous work of the composition  $(\text{NH}_4)_2\text{S}_2\text{O}_8$  for some of the particles lends credibility to the involvement of photochemical reactions

in producing the large particles. In this compound the two  $\text{SO}_4$  units are linked by an ozone-type bond between two oxygen atoms.

The evidence of JUNGE *et al.* (1961) that the large particles have peak number concentrations in the vicinity of 25 km altitude tends to preclude the troposphere as their source. Furthermore, as has been pointed out in the previous section, the size distribution function of stratospheric particles as shown in Fig. 5, cannot be explained on the basis of a steady-state coagulation process. Such a process would tend to fill in the valley between two peaks. It is therefore concluded that the size distribution function deduced in this work (Fig. 5) supports the concept of production in the stratosphere of the large particles. The most likely method of production is a series of chemical reactions among which is the photochemical oxidation of  $\text{SO}_2$ . The rate of reaction is fast compared to coagulation. A portion of the stratospheric Aitken nuclei provide the centers for these reactions. This latter statement is consistent also with the findings of MOSSOP (1965) that insoluble inclusions of the order of 0.04 micron radius were in each large sulfate particle.

The shape of the size distribution function for  $0.3 < r < 1.0$  micron might be explained by a quasi-stationary state of condensation and sedimentation processes, though this is conjectural at present.

## 9. Acknowledgment

The author gratefully acknowledges helpful discussions with Prof. C. E. Junge and the performance of experimental measurements by Mr. Douglas Hallgren of Ernest F. Fullam, Inc., and Mr. Rexford D. Sherwood of Isotopes, Inc.

## REFERENCES

- CADLE, R. D., and POWERS, J. W., 1965, Some aspects of atmospheric reactions of atomic oxygen. CACR Symposium on Atmospheric Chemistry, Circulation and Aerosols, Visby, Sweden 18-25 August 1965.
- FEELY, H. W., DAVIDSON, B., FRIEND, J. P., LAGOMARSINO, R. J., and LEO, M. W. M., 1963, Ninth Quarterly Report on Project Stardust, U.S. Department of Defense, Report DASA-1309.
- FRIEND, J. P., FEELY, H. W., KREY, P. W., SPAR, J., and WALTON, A., 1961, The high altitude sampling program. U.S. Department of Defense, Report DASA-1300.
- JUNGE, C. E., 1961, Vertical profiles of condensation nuclei in the stratosphere. *J. Meteor.*, **18**, p. 501.
- JUNGE, C. E., CHAGNON, C. W., and MANSON, J. E., 1961, Stratospheric aerosols. *J. Meteor.*, **18**, p. 81.
- JUNGE, C. E., and MANSON, J. E., 1961, Stratospheric aerosol studies. *J. Geophys. Research*, **66**, p. 2163.
- JUNGE, C. E., 1965, The formation of the stratospheric sulfate layer. CACR Symposium on At-



- mospheric Chemistry, Circulation and Aerosols, Visby, Sweden, 18-25 August 1965.
- MOSSOR, S. C., 1964, Volcanic dust collected at an altitude of 20 km. *Nature*, **203**, p. 824.
- MOSSOR, S. C., 1965, Stratospheric particles at 20 km altitude. *Geochim. et cosmochim. Acta*, **29**, p. 201.
- NEWKIRK, G., and EDDY, J. A., 1964, Light scattering by particles in the upper atmosphere. *J. Atmos. Sci.* **21**, p. 35.
- RANZ, W. E., and WONG, J. B., 1952, Impaction of dust and smoke particles on surface and body collectors. *Ind. Eng. Chem.*, **44**, p. 1371.

# СВОЙСТВА СТРАТОСФЕРНЫХ АЭРОЗОЛЕЙ

Работа посвящена изучению химического состава, распределения по размерам и концентрации аэрозолей в стратосфере. Показано, что частицы в стратосфере состоят из сульфата аммония и перекиси сульфата аммония и могут также содержать связанную воду.

Было найдено, что средняя функция распределения частиц по размерам является приближенно лагарифмом нормального распределения для частиц радиуса от 0,1 до 1,0  $\mu\text{к}$ . Концентрация частиц являлась переменной, но как правило порядка 0,05  $\text{cm}^{-3}$  на

высоте 18 km. Соответствующая массовая концентрация имеет порядок  $10^{-3} \text{ pg cm}^{-3}$ .

Из комбинации, полученной функции распределения с оценками функции распределения по размерам ядер в стратосфере следует, что полное распределение частиц материи в нижней стратосфере может быть бимодально, т. е. иметь два максимума. Это показывает, что должны иметь место другие процессы помимо коагуляции, если частицы радиусом  $< 0,1 \mu\text{к}$  дают начало частицам радиуса  $> 0,1 \mu\text{к}$ .