

The Size and Number Distribution of Aerosols in the Continental Troposphere

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ABSTRACT

A series of 22 aerosol collections made during the year 1966 at altitudes to 10 km in the vicinity of Scottsbluff, Nebr., have been analyzed. Several additional samples were taken at other locations in the central United States when low tropopause conditions permitted collecting in the lower stratosphere. A few additional measurements were made over Limon, Colo. The sampling device was a single stage impactor mounted in a Beech Queen Air aircraft. Particle size and number distributions were determined by direct counting from photomicrographs. The collecting and counting techniques are described and the experimental results presented. Systematic changes were observed in the aerosol distribution function with altitude in which both the large and small particulates diminish with height. Short term variations in the size and number distribution, seasonal changes, and correlations with various meteorological parameters were investigated. Characteristic aerosol distribution functions for tropospheric and lower stratospheric air were found on occasions of low tropopause. Correlations of the total number of large and intermediate size particles with relative humidity and a more or less constant mixing ratio of intermediate sized particles with altitude suggest the importance of cloud modification processes in determining the aerosol distribution function.

1. Introduction

The size and number distribution of the atmospheric aerosol near the ground has been the subject of a large number of investigations including those of Junge (1955), Pasceri and Friedlander (1965), and Clark and Whitby (1967). Although some information is available for stratospheric altitudes from the work of Chagnon and Junge (1961), and Mossop (1965), except for measurements by Fenn *et al.* (1965), the troposphere remains largely unexplored. In attempting to generalize the observed size and number distribution, Junge (1957) proposed that a constant log-volume distribution, presumed to result from contributions from a large number of diverse sources, would explain the observed constant slope of the distribution function. For large and giant particles, Podzimek (1965) adopts a different form of the distribution function which is also based on statistical considerations. Much attention also has been given to the sedimentation-coagulation theory of Friedlander (1960) who derived theoretically essentially the same distribution proposed by Junge.

Recently, however, Junge (1964) has made extensive model computations in which the coagulation equations are solved numerically and in which the effects of evaporation-condensation, rain-out, sedimentation, and other physical and chemical processes are included. Although this work is incomplete it seems to give promise of providing an explanation of the observations based on a realistic model of the atmospheric processes that affect the particle size distribution. The present

work was undertaken in the expectation that information on the aerosol content of the troposphere over the mid-continent would be useful to this general theory and also in optical computations requiring knowledge of the aerosol distribution function. The location chosen for our measurements was Scottsbluff, Nebr., where 22 sets of samples at several altitudes to 9.1 km were obtained over the period from February 1966 through January 1967.

2. Instrumentation

The sampling device was a single-stage jet impactor¹ designed for an altitude of 40,000 ft and used by Chagnon and Junge (1961) for their balloon collections. We provided a removable end plate to facilitate changing the microscope slide during flight. The original calibration made by General Mills Corporation employed an optical scattering method to determine the test aerosol concentration. Although Chagnon and Junge's procedures were followed as closely as possible, we felt that it was desirable to recalibrate using a direct method. The necessary measurements were made for us by Dr. K. Spurny² of the Czechoslovak Academy of Science using techniques which he has developed for

¹ A set of calibrated balloonborne impactors were made available by Mr. C. W. Chagnon of the Air Force Cambridge Research Laboratories, Bedford, Mass.

² The impactor efficiency measurements were made for us while Dr. Spurny held a visiting scientist appointment at the National Center for Atmospheric Research, September 1966 to September 1967.

producing radioactive test aerosols. The method involves the production of particles whose size can be accurately controlled and measured using the superior resolving power of the electron microscope, thus avoiding the necessity of diffraction corrections. The experimental details are described by Spurny and Lodge (1968). The efficiency was determined by comparing, with a Geiger counter, the radioactivity of the particles deposited by the impactor with that of the particles collected on an efficient membrane filter. The resulting efficiency curve is shown as Fig. 1 along with the original calibration by General Mills. We feel that the radioactive technique is superior to the method used by General Mills since it gives a direct measure of the material deposited beneath the jet and that the reason for the difference between the two calibrations probably is due to deposition of particles within the device. This effect would cause the higher efficiencies observed by General Mills.

Ranz and Wong (1952) describe the theoretical efficiency of a jet impactor by the use of a nondimensional parameter

$$\Psi^{\frac{1}{2}} = (C\rho_p V / 18\eta d_j)^{\frac{1}{2}} d_p, \quad (1)$$

where η is the viscosity of air, V the air velocity at the jet, ρ_p the density of the particle, d_p the diameter of the particle, d_j the width of jet orifice, and C the Cunningham correction

$$C = 1 + 2\lambda/d_p [1.23 + 0.41 \exp(-0.44d_p/\lambda)], \quad (2)$$

where λ is the mean free path of air molecules. The form of $\Psi^{\frac{1}{2}}$ found from the calibration measurements and put into the above equations provides a means of obtaining the impactor efficiency for other values of the parameters, which in our case were used to correct the impactor efficiency for changes in altitude.

Eq. (1) applies strictly to the particles deposited directly under the jet orifice. However, in many of our collections we observed a diffuse deposit surrounding the central band where this "ring" seems to be due to turbulent effects which are not taken into account by the theory. It is discussed at some length by Roeber (1957) who found that increasing the thickness of a layer of binding agent applied to the collecting surface would suppress or prevent formation of the ring. However, we made a number of tests in which several times the thickness of the film of silicone oil found to be efficient by Junge failed to eliminate it. Therefore, in our work the size and number distribution of the particles in the ring and its geometry were used to correct the distributions which we determined from the central deposits. We feel, furthermore, that the radioactive calibration technique allows this effect to be accounted for satisfactorily.

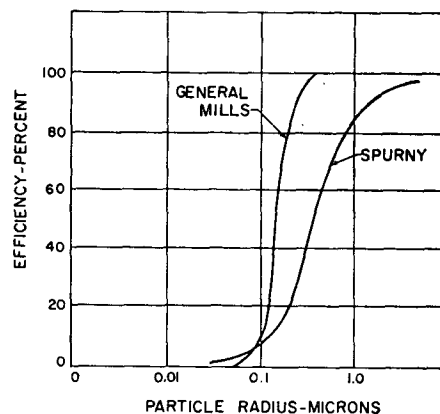


FIG. 1. Impactor efficiency as a function of particle radius.

3. Aircraft installation and particle size analysis

The impactor was installed in the cabin of a Beech Queen Air aircraft which has an altitude capability of approximately 10 km. The air intake, mounted above the fuselage, consisted of a long sharp-edged nozzle which, at the average aircraft speed, provided isokinetic flow that was maintained by a combination of ram pressure and suction. The exhaust air was vented below the fuselage and constant volume flow was obtained by manual adjustment in flight. The impactor collecting surface, a microscope cover slip mounted on a standard microscope slide, was carefully cleaned and coated with filtered silicone oil. All operations were performed in a clean room where the prepared slides were loaded into a sealed carrying case provided with a membrane filter to allow pressure adjustment with changes in altitude. The slides were exposed to the cabin environment only during insertion and removal from the impactor. Control slides were always processed and carried along with the exposed ones but little contamination was encountered and no correction for it was found to be necessary. Upon return of the exposed slides to the laboratory clean room, the cover slips were reversed and photomicrographs were made using Polaroid *P/N* or Panatomic 4×5 cut film. Bright field phase contrast at a magnification of about 800 diameters was used in all cases. With each set of samples a photomicrograph of a stage micrometer provided an accurate scale for subsequent enlargement to a standard magnification of 2500×.

The particle images on the photographic prints were measured and counted using the Zeiss TGZ3 particle counter. This instrument produces calibrated circular optical images which, at the magnification chosen, allowed particle classification into 48 logarithmic intervals ranging from 0.130 to 5.52 μ . The image diameters were checked with an accurate scale and microscope. The data obtained from the particle size measurements were punched on cards and a computer program written for making the corrections for variation of

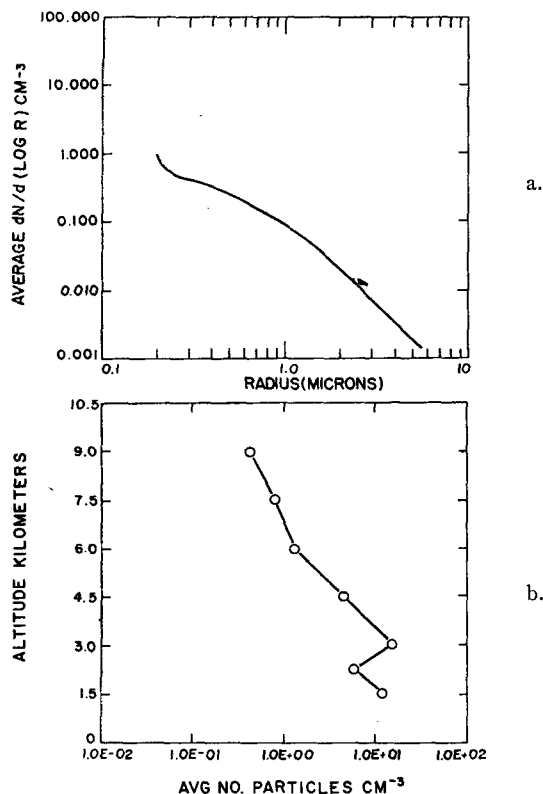


FIG. 2. Average $dN/d(\log R)$ [cm^{-3}] vs particle radius for all dates and all altitudes, a., and average number of particles (cm^{-3}) with radii $0.13 \leq R \leq 5.52 \mu$ vs altitude of collection for all dates and all altitudes, b.

impactor efficiency with altitude, air flow and time of collection, as well as the various geometric, optical and photographic corrections necessary to combine particle counts from the center band and the diffuse ring. Curves of the distribution function $dN/d(\log R)$ and particle number vs altitude were produced as direct computer output.

The final optical calibration was made from measurements on polystyrene latex spheres deposited on microscope slides. We recognize the inadequacy of polystyrene latex particles as calibration standards for the atmospheric aerosol. However, we felt that an estimate of the diffraction effect for particles with diameters $< 0.5 \mu$ was necessary. The correction factors obtained ranged from about 1.9 at 0.13μ and 1.3 at 0.516μ to 1.04 for 1.05μ particles. These values include a correction of approximately 10% for the spot size of the TGZ3. Since even 0.5μ particles appear to have radii which are about 30% larger than the true radii, our data will show an apparent shift to larger particle sizes which is not completely determinable because the effect is strongly dependent on particle composition. All of our results, however, have been adjusted to take the diffraction effect into account using corrections for diffraction obtained by photographing and measuring polystyrene latex spheres with accurately known diam-

eters in five size ranges from 0.234 to 1.099μ . Although the limitations of optical microscopy make for some uncertainty in the true particle number in the small size range, we believe that the general form of our distributions will not be seriously affected. We have taken great pains to obtain uniformity in all of our experimental techniques and feel that our relative values should be quite accurate.

The efficiency of the impactor is quite low for particles with radii $\leq 0.2 \mu$ and it was necessary to correct our data accordingly. Although we made a somewhat arbitrary decision to cut off the distributions where the calculated efficiency was less than 30%, it is possible that in some cases the sharp increase in slope for the smallest particles may be fictitious. It should be noted also that at the higher altitudes this effect tends to be enhanced because of increased impactor efficiency.

4. Sampling procedure

Collections were made in sets of eight altitudes, with the lowest about 0.3 km above the ground to a maximum of 9.1 km. In the first half of 1966 measurements were made approximately three times each month and, except for the month of August, at least once per month during the rest of the year. The reasons for selection of the collecting site at Scottsbluff were 1) its convenience to our laboratory at Boulder, Colo.; 2) its geographical location which is approximately midway between the paths of cyclonic systems from the arctic and the Gulf; and 3) the altitude capability and range of the aircraft. It is remote from sources of industrial pollution, and is believed to be sufficiently far from the mountains to minimize their influence. On all missions the aircraft were directed to maintain constant altitude, to fly in an approximately circular path, and to avoid clouds and obvious sources of local contamination. The flight path was chosen to be nonintersecting, and, at the measured wind speeds, we feel that self-contamination from the aircraft was unlikely. All flights were in clear weather with a duration of 1 hr except at the lower altitudes where the sampling time was 30 min. During the day of the flights two standard radiosondes were flown, one of which carried an ozone-sonde of the Brewer type.

5. Aerosol size distribution in the troposphere

For convenience in comparing our results with those in the literature we have chosen to present our data in terms of logarithmic distribution function $dN/d(\log R)$ [cm^{-3}], where N is the number of particles cm^{-3} smaller than R and R the particle radius.

Fig. 2a shows this average logarithmic distribution function vs particle radius for all of our approximately 200 measurements at altitudes from 1.5 to 9.1 km during the year 1966. This average curve has a slope of about -2.3 and is probably representative of the aerosol population below 3.0 km. High particle concentrations

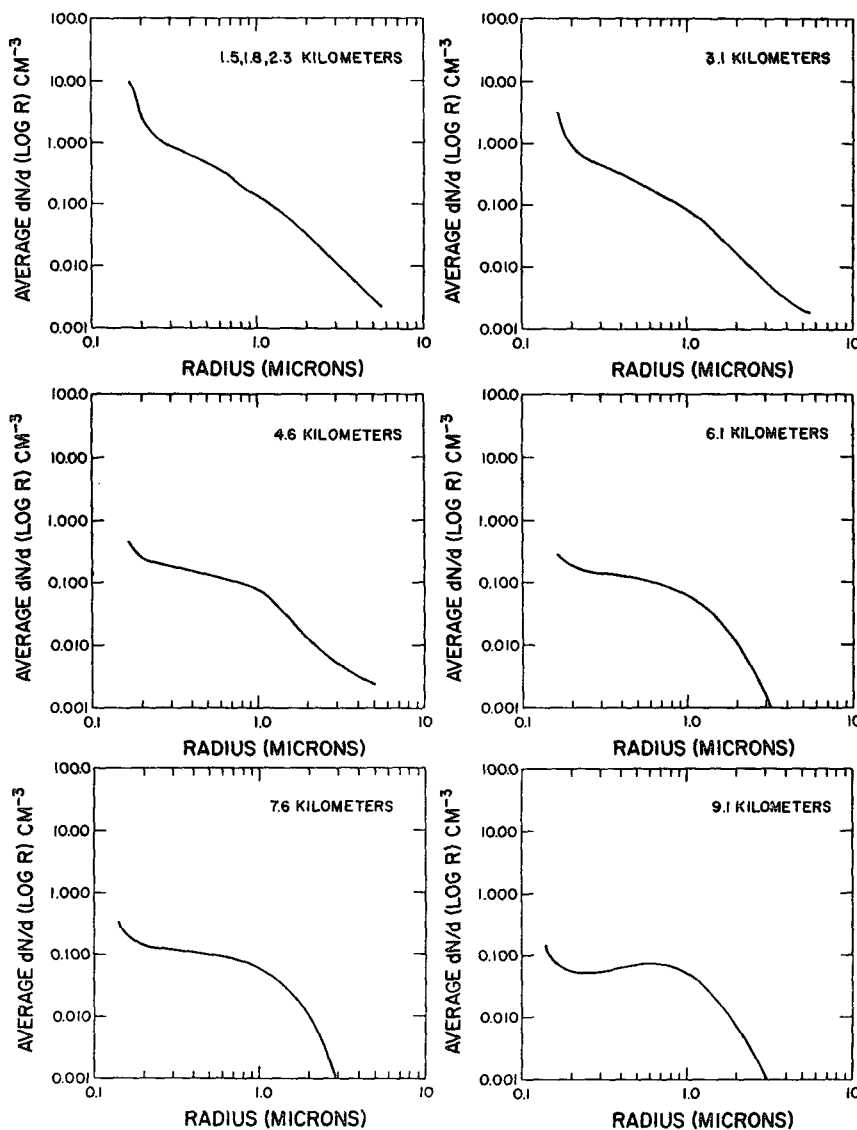


FIG. 3. Average $dN/d(\log R)$ [cm^{-3}] vs particle radius for altitudes of 1.5, 1.8 and 2.3 km combined, and for 3.1, 4.6, 6.1, 7.6 and 9.1 km.

at the lower altitudes tend to weight the average values more heavily than the lower concentrations encountered at high altitudes.

Fig. 2b gives the average total number of particles with radii between 0.13 and 5.5 μ as a function of altitude for the same measurements as in Fig. 2a. The total particle concentration in this size range decreases from about 11.0 cm^{-3} at 1.5 km to about 0.04 cm^{-3} at 9.1 km. These values agree with the estimates of Chagnon and Junge (1961) which were based mainly on optical scattering data. In reviewing the available literature they obtained values of 3.0–10.0 cm^{-3} at 1.5 km and their own balloonborne impactor measurements gave 0.01 to \sim 0.03 cm^{-3} at an altitude of 11 km.

In Fig. 3 average curves of $dN/d(\log R)$ [cm^{-3}] vs

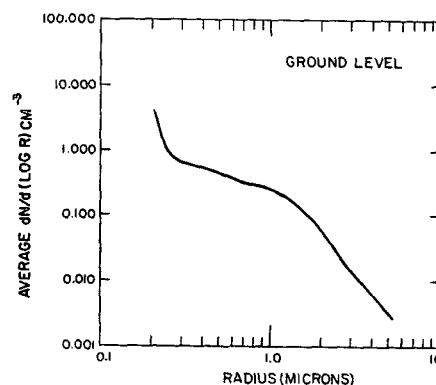


FIG. 4. Average $dN/d(\log R)$ [cm^{-3}] vs particle radius at ground level.

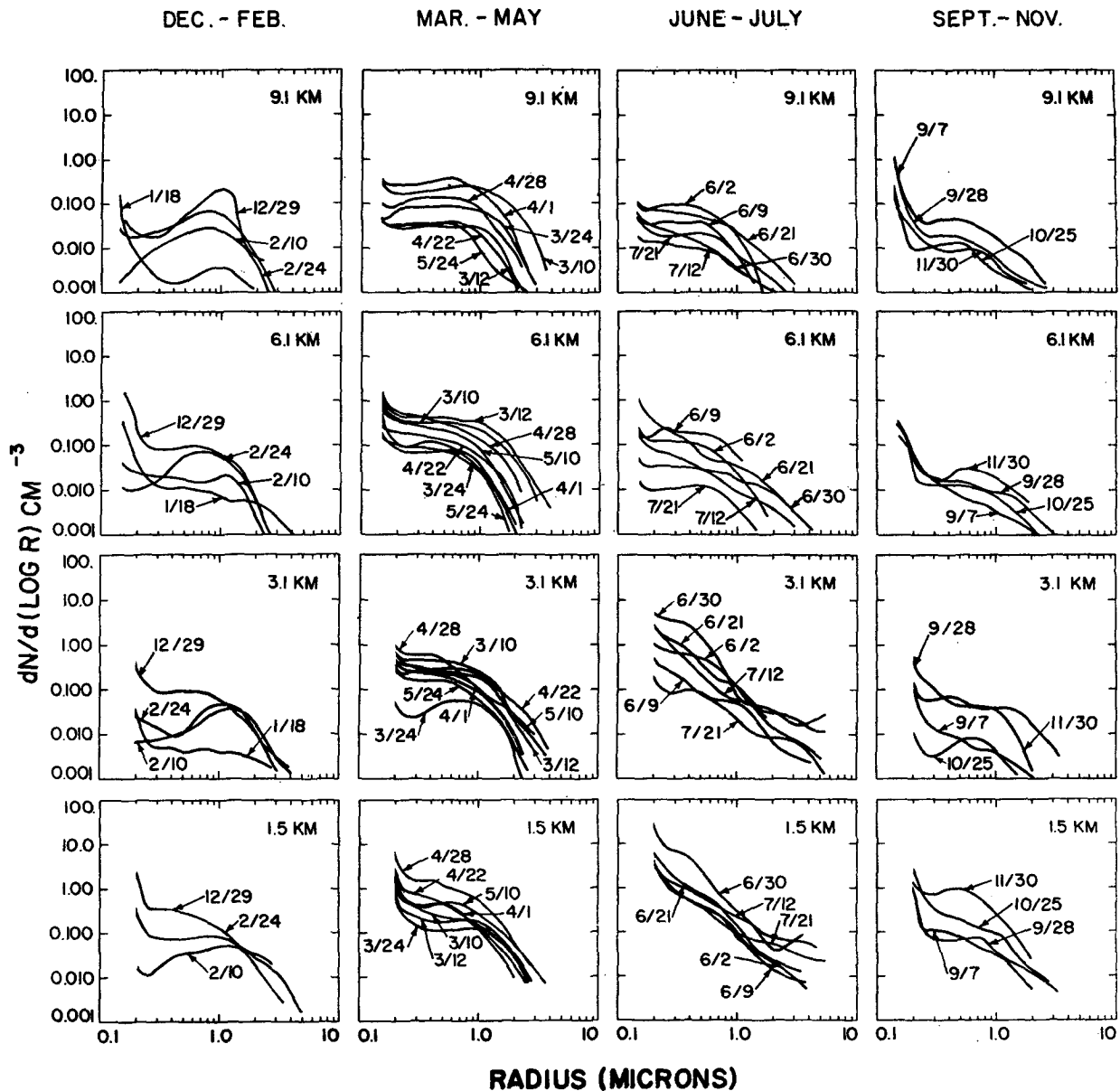


FIG. 5. Individual $dN/d(\log R)$ [cm^{-3}] vs particle radius for four altitudes arranged by season.

particle radius are shown for the combined altitudes 1.5, 1.8 and 2.3 km and for 3.1, 4.6, 6.1, 7.6 and 9.1 km. Each represents the average of collections made on 22 days during 1966-67 at Scottsbluff. The altitudes indicated are pressure altitudes, i.e., the 1.5-km level is approximately 0.6 km above the ground. The data from the three lower altitudes were combined because the distribution curves were so nearly identical; the low altitude curve, therefore, is the average of 66 individual measurements. In obtaining it, $\sim 6 \times 10^4$ particles were counted, while for each of the remaining altitudes $\sim 2 \times 10^4$ particles were counted. For comparison, an average ground level curve is shown in Fig. 4 although

it is the average of only eight measurements made in August 1967 and therefore probably is not as representative as the distributions of Fig. 3. There seems to be a systematic change of slope with altitude, from about -2.0 at the lower levels, to distributions in which for the larger sizes the slope approaches -3.0 accompanied by a decided levelling off in the range of 0.2 - 1.0μ . The most obvious characteristic seems to be compression of the distributions obtained at the higher altitudes by reduction of both the largest and smallest sizes.

The radius intervals which we used for particle size classification were separated by an average of 0.04μ for particle radii $< 1.0 \mu$ and 0.23μ for radii $> 1.0 \mu$.

With this resolution none of the distributions showed the presence of "holes" as reported by Fenn (1964) although there is some structure in individual curves. Many of our distributions are similar to those reported by Fenn for the Texas Prairie area.

A significant characteristic of our data seems to be the wide variation in the size and number distributions at all altitudes over the period of measurement. This feature is not obvious from the average values of Fig. 3, but is evident in Fig. 5 which shows all of the individual curves for four altitudes arranged by season. At lower altitudes during the summer the size distributions tend toward greater uniformity and conform more closely to the "standard form" with -3 slope. It should be noted that data for August are missing since aircraft scheduling problems prevented our making measurements during that month. The variability in concentration appears to be greater during the winter, with the winter troposphere exhibiting lower average values at all altitudes.

6. Stratospheric size distributions

Some of the higher altitude samples were taken when standard maps of tropopause height indicated that the altitude of collection was as much as several thousand feet above the tropopause. The presence of stratospheric air was confirmed in each case by high concentrations of the radioactive stratospheric tracers Cs-137 and Be-7. The distribution function for the average of these six collections made at 9.1 km under the above conditions is shown in Fig. 6a with dates, locations and tropopause heights given in Table 1. Fig. 6b shows the individual logarithmic distribution functions for the same set of six observations. In stratospheric air the distribution peaks in the vicinity of particle radius 0.5μ . Tropospheric air as represented by the average of 21 Scottsbluff samples collected at the same altitude do not show this peak. However, individual distribution curves for stratospheric samples seem to show no greater variability than do winter and spring samples taken at the same altitude at times when there is no indication of stratospheric air. It is possible that the observed average distribution may be characteristic of the lower stratosphere but because of the variability and relatively

TABLE 1. Aerosol collections that coincide with meteorological evidence of low tropopause and high stratospheric radioactivity at 9.1 km.

Date	Tropopause height (km)	Location
10 Feb. 1966	7.3	Scottsbluff, Nebr.
15 Mar. 1968	7.9	Scottsbluff, Nebr.
16 May 1968	7.3	Scottsbluff, Nebr.
17 Mar. 1966	8.6	Rapid City, S. D.
4 Apr. 1966	7.9	Garden City, Kan.
20 Mar. 1968	7.3	El Paso, Tex.

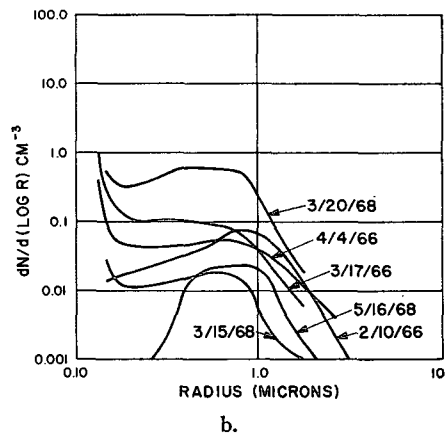
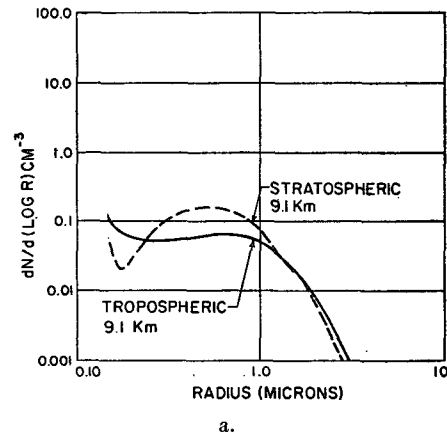


FIG. 6. Average $dN/d(\log R)[\text{cm}^{-3}]$ vs particle radius for collections made in stratospheric air and in the troposphere at 9.1 km, a., and $dN/d(\log R) [\text{cm}^{-3}]$ vs particle radius for individual collections made in stratospheric air at 9.1 km, b.

small number of samples, our present experimental evidence is probably insufficient for drawing this conclusion.

7. Short-term variations

It has not been possible to carry out extensive measurements to determine the short-term variability of the particle size distributions. However, at Scottsbluff we were able to make sets of collections two days apart. The distributions obtained at 3.1 and 7.6 km are shown in Fig. 7a, and for 6.1 and 9.1 km in Fig. 7b. The curves for 3.1 and 6.1 km are similar on both days, but at 7.6 km there seemed to be a considerable decrease in large particles on 12 March with little change in the number of particles with radii $\leq 1.0 \mu$. At 9.1 km a small decrease in large particles was accompanied by an overall decrease by a factor of 10 for the entire distribution. Except for the highest altitude, however, the distributions were not very different.

The data of Table 2 give some indication of the variability in the total particle concentration on the scale of hours as determined from measurements made 6

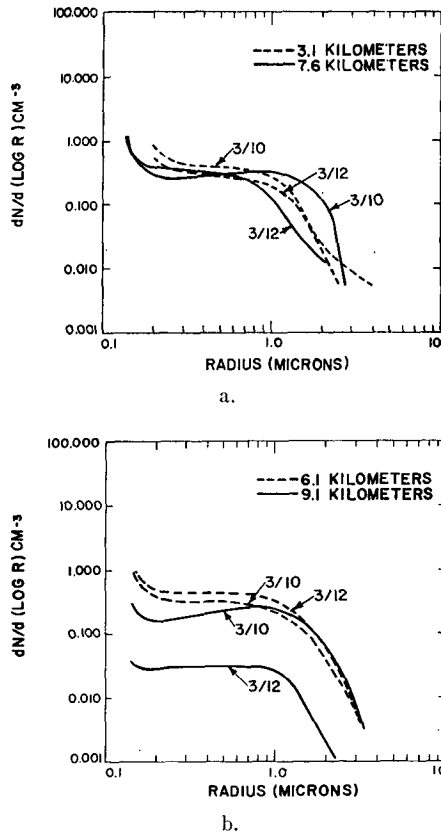


FIG. 7. Average $dN/d(\log R)$ [cm^{-3}] vs particle radius at 3.1 and 7.6 km for collections made two days apart, a., and average $dN/d(\log R)$ [cm^{-3}] vs particle radius at 6.1 and 9.1 km for collections made two days apart, b.

September 1967 near Limon, Colo. Three samples were taken for successive 60-min periods at altitudes of 4.9, 6.7 and 8.5 km using two aircraft. It appears that variations by a factor of at least 4 may not be uncommon. These experiments do not, of course, differentiate between temporal and spatial variations.

8. Comparison of optical and direct methods

For several samples taken at 8.5 km altitude it was possible to compare the direct particle concentrations

TABLE 2. Short-term variations in total particle concentration for particles with radii in the range of 0.13 – 5.5μ on 6 September 1967 over Limon, Colo.

Time (GMT)	Altitude (km)	Particle concentration (cm^{-3})
2340	4.9	0.14
2440	4.9	0.056
2540	4.9	0.047
1848	6.7	0.087
1948	6.7	0.11
2048	6.7	0.027
1906	8.5	0.26
2006	8.5	0.065
2106	8.5	0.11

TABLE 3. Comparison of particle concentration for particles with radii in the range of 0.5 – 5.0μ as measured at 8.5 km using a single stage impactor and a Royco, Inc., optical counter.

Date	Time (GMT)	Optical counter (particles cm^{-3})	Impactor (particles cm^{-3})
9-6-67	1906	0.059	0.034
9-6-67	2006	0.063	0.035
9-6-67	2106	0.055	0.044

obtained from the impactor with in-flight measurements using a Royco, Inc., optical particle counter. These data are given in Table 3 for particles $0.5 \leq R \leq 5.0\mu$, the range of particle radii for which the optical counter was adjusted. In this size range and for the limited comparisons that were possible, the two methods agreed within an average factor of 1.6. It may be noted that the sample labeled 2006 seems to have a much smaller proportion of small particles than either of the other two.

9. Correlations with atmospheric parameters

Trajectories of the air masses which were sampled have not been analyzed. However, a statistical study was performed in which correlations between the aerosol data, the radiosonde and ozonesonde data, and concentrations of certain cosmogenic and fission product radioactivities were computed. Ozone concentrations were derived from analyses of ozonesonde data using the procedures of Hering and Dutsch (1965).

The air concentrations of the radioactive, cosmic-ray-produced isotope Be-7 and fission-product Cs-137 were obtained from gamma spectral analyses of filter collections made simultaneously with the impactor samples.³ Tritium was determined from low level beta measurements of atmospheric water vapor also collected at the same time.⁴ Table 4 gives the correlation coefficients for the above parameters with the total particle concentration (cm^{-3}) for all altitudes and all dates. The letters NS indicate a coefficient not significantly different from zero at the 2σ confidence level. It appears

TABLE 4. Correlation coefficients for the total particle concentration with meteorological and air chemistry parameters measured simultaneously. Data for all altitudes and all dates were used.

Parameter	Correlation coefficient
Wind direction	-0.27
Wind speed	-0.20
Relative humidity	NS
Temperature	+0.40
Total tritium	+0.39
Be-7	NS
Cs-137	NS
Ozone	NS

³ Private communication from Dr. J. P. Shedlovsky, to be published.

⁴ Private communication from Dr. D. Ehhalt, to be published.

TABLE 5. Correlation coefficients for the particle mixing ratios for four particle size ranges with meteorological and air chemistry parameters measured simultaneously. Data for all altitudes and all dates were used.

Particle radius (μ)	Wind direction	Wind speed	Relative humidity	Temperature	Tritium units	Be-7	Cs-137	Ozone
0.13-0.52	-0.25	-0.19	NS	+0.38	NS	NS	NS	NS
0.52-1.01	-0.21	NS	NS	+0.24	NS	NS	NS	NS
1.01-1.96	NS	NS	+0.25	NS	NS	NS	NS	NS
1.96-5.52	-0.32	-0.16	+0.17	+0.29	NS	NS	NS	NS

that low wind speed and east winds have a tendency to produce higher total particle concentrations. It may be that the plains to the east of Scottsbluff are a better source of soil aerosols than the relatively barren mountains to the west. Positive correlations with temperature and total tritium probably indicate only that these parameters decrease with altitude rather than the existence of a specific physical relationship.

Table 5 presents the correlation coefficients for four particle size groups with the same parameters as in Table 4. In this case the data were expressed in terms of mixing ratios. The same correlations seem to be significant except that, in addition, the larger particles are positively correlated with relative humidity. Additional correlation coefficients computed for the available data at individual altitudes and with season gave significant values only at 4.6-6.1 km where the concentration of Be-7 and Cs-137 (tracers for stratospheric

air) appeared to be related to the particulate concentration. We have no indication, however, that this is the case at the highest sampling altitude where large concentrations of these isotopes seem more probable.

10. Seasonal variations and altitude distribution of total particle number

The total number per unit volume of air of particles in the size range of our measurements seems to be strongly dependent on the season. In Fig. 8 the average number of particles per unit volume with radii 0.13-5.53 μ for all altitudes up to 9.1 km is shown for each date of collection. The particle concentration begins to increase from about 1.0 cm^{-3} in March to about 6.0 cm^{-3} in early June. A maximum of about 30.0 cm^{-3} was reached in late June and by September the concentration had returned to about 1.0 cm^{-3} . Again no measurements are available for August.

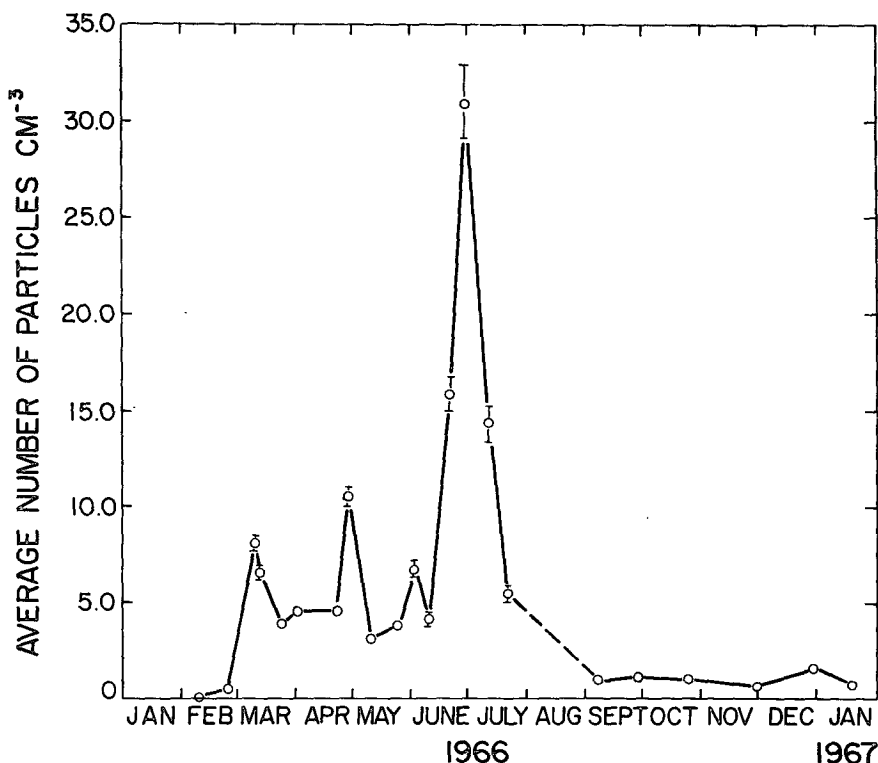


FIG. 8. Average total number of particles with radius $0.13 \leq R \leq 5.52 \mu$ for altitudes 1.5-9.1 km vs date of collection.

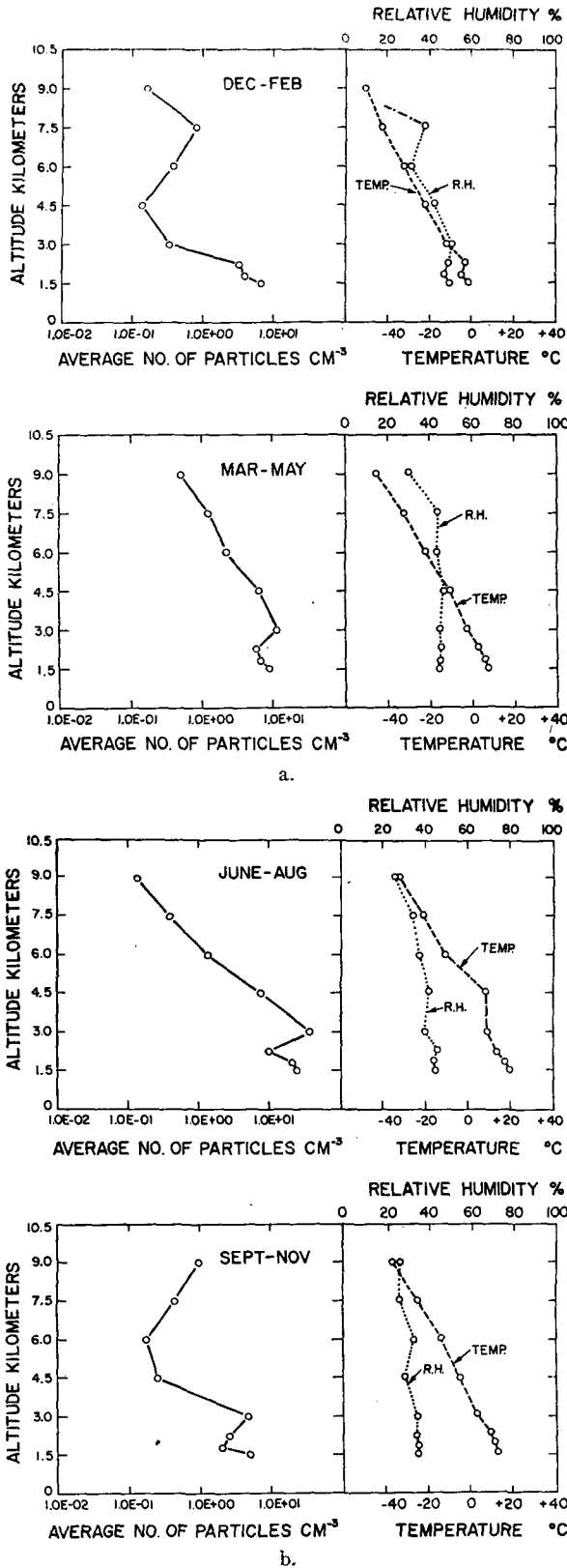


FIG. 9. Average total number of particles with radii $0.13 \leq R \leq 5.52 \mu$ and average temperature and humidity as a function of altitude for December-February and March-May, a., and for June-August and September-November, b.

Seasonal curves of the average particle concentration vs altitude are shown in Figs. 9a and 9b from which it is evident that summer is the period of highest concentration. This seasonal trend is confirmed by individual inspection of the entire collection of 22 curves of particle number vs altitude. The increase at about 7.6 km seems to be characteristic of the fall and winter. The particle concentration at the higher altitudes decreases somewhat during the spring and summer while at the lower levels there is a marked increase. In general, it appears that the aerosol population has two distinct seasonal regimes, viz., summer and winter.

Curves of average temperature and average relative humidity as a function of altitude obtained from radio-sonde data taken at Scottsbluff simultaneously with the aerosol collections are also shown in Figs. 9a and 9b. The particle concentration does not seem to be closely related at any season to the stability or vertical exchange at least as indicated by the accompanying average temperature profiles. Except for the December-February curves there also does not seem to be an obvious relationship between average relative humidity and particle concentration. In the winter months there appears to be a reduction in the number of particles with very low humidity. It should be noted that in all seasons the average humidity was below 75%, the value at which sea salt particles are generally assumed to be liquid.

The total mass of particles with radii in the ranges $0.13-0.52 \mu$, $0.52-1.0 \mu$, $1.0-2.0 \mu$ and $2.5-5.5 \mu$ was computed assuming spherical particles of density 2.0. These

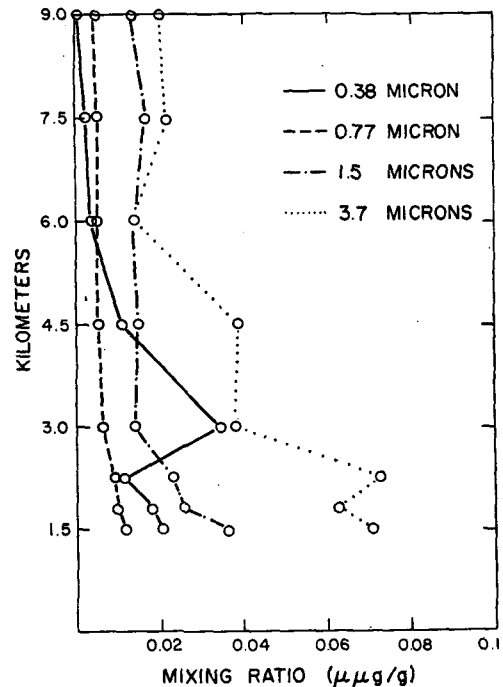


FIG. 10. Average particle mixing ratio ($\mu\text{g gm}^{-1}$ of air) vs altitude for the particle size fractions of mean size 0.38, 0.77, 1.5 and 3.7μ .

data were averaged, converted to mixing ratios and plotted vs altitude in Fig. 10 where each particle group is identified by its median radius in microns. From these curves it may be seen that the mass of particles of radius 0.5–2.0 μ decreases very little from 1.5 to 9.1 km while the relative decrease with altitude is much greater for both the larger and smaller size fractions. At 3 km the minimum for large particles appears to coincide with a maximum for small particles.

11. Summary and conclusions

Our results for the aerosol size and number distributions near the earth's surface seem to be more or less in agreement with previous measurements, i.e., the logarithmic distribution function can be described roughly as having a constant slope of -2 to -3 when plotted as a function of particle radius. At altitudes ≥ 3 km, however, we observe a systematic decrease in slope for the particle size range 0.1–1.0 μ . At the highest altitudes there is a marked reduction in both the large and small particles. The above observations are consistent for higher altitudes and presumably older aerosols, with the idea that coagulation tends to reduce the population of small particles, while sedimentation and capture by rain removes the larger particles. Processes such as condensation, evaporation and other modification phenomena within clouds therefore have a higher probability of occurrence in the intermediate size range.

The decrease which we observe in particle number with height confirms, by direct measurement, estimates made previously which were based on optical observations. To the limit of our particle size resolution (0.04–0.23 μ) we do not observe "holes" in the distributions. We find that on the average the rate of decrease of the total aerosol mass is not uniform for all particle sizes. It appears that the mixing ratio of the intermediate size fraction (0.5–2.0 μ) is nearly constant for all altitudes to 9.1 km. The decrease with altitude of this quantity for larger and smaller sizes follows the temperature structure. These observations again suggest the importance of modification processes within clouds.

Individual observations indicate considerable variability in the size distribution on the scale of weeks at all altitudes. We observe a marked seasonal effect in both size and number, which, from visual appearance and the increase in the number of larger particles, seems to be due to an enhanced biological component appearing in early summer. The increase in number of spores, pollen, and insect and plant fragments is quite apparent at this time. In collections made two days apart the size and number distributions at the lower altitudes were similar. The largest difference was found at altitudes of 7–9 km. An effort to observe variations over periods as short as 30 min gave indications of fluctuations in total aerosol number by a factor of 4. It is not clear whether the inhomogeneities are primarily temporal or spatial in nature.

Computed correlation coefficients between the particle concentration, wind speed and wind direction indicate that higher concentrations are associated with east winds and low wind velocities. The larger particles seem to correlate positively with relative humidity while no significant correlations with tracers for stratospheric air could be established. On the whole the correlations with meteorological parameters are about what might be expected from other qualitative considerations, in which the larger particles are generally thought to be important in condensation and to be affected by growth processes at high humidity. The presence of relatively more or less stratospheric air does not seem to be correlated with particle number.

The aerosol distribution function for stratospheric air seems to follow the general pattern of decreasing relative concentration of both small and large particles with altitude. The available data, however, do not clearly indicate a characteristic distribution function associated with stratospheric air masses.

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