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Size Distributions of Stratospheric Aerosols  
and their Variations with Altitude and Time

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(Manuscript received

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# Size Distributions of Stratospheric Aerosols and their Variations with Altitude and Time

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## ABSTRACT

↙ A description is given of the variation with altitude and time of the size distributions and concentrations of stratospheric particles collected with impactors carried on 35 Australian balloon flights from latitude 34<sup>deg</sup> S. in the five years May 1969 to April 1974. Seasonal variations were found to be confined mainly to the lower stratosphere, where a winter maximum of small particles was followed by a spring maximum of large particles. Longer-term trends showed only one conspicuous feature, a decrease in concentration of about one order of magnitude in 1971. Former concentrations were restored by the beginning of 1972 by an increase commencing in late August 1971. The size distributions as a function of altitude were very similar to those obtained by Junge and his colleagues with balloon-borne equipment in the U.S.A. 10 years earlier and with those obtained in Wyoming, U.S.A. in 1972. The main features are modal diameter around 0.2  $\mu\text{m}$  at the top of the layer which falls to less than 0.1  $\mu\text{m}$  at lower levels. There is also some indication of an inflection in the lower-level curves at about 0.5  $\mu\text{m}$  diameter.

## 1. Introduction

It is now well established that the presence of aerosols in the stratosphere leads to a net stratospheric warming and a cooling at the earth's surface. The magnitude of the effect depends on the altitude at which the particles are located, their concentration and size distribution and their properties relevant to scattering and absorption of radiation. It is thought that aerosols put into the stratosphere by major volcanic eruptions can significantly affect climate. Conclusive empirical evidence that this is so has been difficult to obtain but studies such as Lamb's (1970) certainly suggest that it is.

Because widespread climatic change is potentially of great importance it is essential that its causes be understood and the possible contributions from the stratospheric aerosol be assessed. To understand this role of particles in the high atmosphere it is necessary to obtain all their relevant properties as a function of altitude and time.

Methods that are commonly used for aerosol monitoring, their advantages and disadvantages are listed below.

(1) Turbidity measurements from the ground. These are relatively easy to make and so provide good continuity. However, the effects of an aerosol on surface temperature depends on the way in which it is distributed in the vertical as well as on its properties, and this information cannot be obtained from turbidity data.

(2) Optical radar. Because it provides a full description of light scattering as a function of altitude, and can give reasonable continuity it is greatly superior to turbidity measurements. It can however tell us nothing about the nature of the particles, and changes in scattering may be due either

to changes in number or size distribution. Thus, interpretation of the results is difficult.

(3) The balloon-borne optical particle counter used by Rosen (1964) for many years and in many places. It cannot readily give the continuity in time that turbidity or optical radar measurements provide and cannot show the nature of the particles, but it does give a detailed record of the number larger than a certain size as a function of altitude.

(4) Collection of particles by a balloon-borne impactor. The method was used by Junge et al. (1961) to investigate simultaneously the size distribution (using an electron microscope), concentration and chemical nature of the stratospheric aerosol. Their work still provides the most complete single description of the properties of the stratospheric aerosol relevant to interpretation of its origin and its role in climatic change. The disadvantages are that electron microscope examination of impaction samples is tedious and time-consuming and the accuracy of concentrations deduced by the impactor is in question when large solid particles are being collected, or when the particles are so small that collection efficiency of the impactor is low.

In spite of these disadvantages, electron microscope examination of impacted aerosol samples has such a clear advantage in terms of available information that we initiated a program at the end of 1967 to extend Junge et al.'s (1961) work. The present paper describes the results of the five years May 1969 to April 1974. Results from 1968 experiments have been described by Bigg and Ono (1971) and more detailed results of a parallel investigation in conjunction with the University of Wyoming's optical counter over Wyoming, U.S.A. in 1972 have been described by Bigg (1975). The latter work

showed that the impaction technique gave reasonable agreement with the optical counter on particle concentrations much of the time but that when frozen sulfuric acid particles were present tended to give lower concentrations, particularly in the larger size ranges. There are two probable reasons - one is simply that the impactor is likely to lose some of the irregular chains of spheres that shatter on impact. The other is that the optical counter will describe such a chain as a single particle while in the impactor collections the individual components must be counted unless it is obvious that aggregation subsequent to collection was not a possibility.

## 2. Sampling and data reduction methods

The equipment used is the same as that described in earlier papers. A vacuum pump drew about  $10 \text{ l min}^{-1}$  of air through a 1 mm diameter impactor nozzle on to electron microscope screens mounted in a circle on a slowly rotating turntable. After retrieval, the screens were shadowed with gold-palladium alloy at an angle having a tangent of  $1/2$ , in order to reveal the height of the particles. Photographs were then taken using a transmission electron microscope, and sizing of the particles was carried out on the negatives with a projection microscope. The assumption on the shape of the particles was based on the outline of their "shadows". A common type of particle had a rounded outline but was considerably flatter than a sphere. It was assumed that such particles were spherical caps and the diameter of the "equivalent sphere" was calculated. The most common particles however consisted of concentric rings of small droplets usually surrounding a somewhat flattened central particle. Collection on copper or on sodium carbonate surfaces showed

the droplet rings to be sulfuric acid and led to the conclusion that the droplet rings were due to the acid in the collected particle having spread out into a monomolecular layer about the non-acid portion and then having taken up water vapor on exposure to higher humidities. On this basis the percentage of sulfuric acid in the particle and the "equivalent spherical diameter" of the whole particle could be calculated. Such particles often appeared to have been frozen aggregates in the atmosphere and some good photographic evidence of this has accumulated.

The estimation of collection efficiencies, using the Ranz and Wong (1952) equations, has been described in the earlier publications. Concentrations were estimated from complete scans with the electron microscope of strips perpendicular to the impaction track. In forming the size distributions 10 equal logarithmic steps per decade of diameter were used and enough strips averaged to provide at least 100 particles but often as many as 1000.

When the stratosphere was "disturbed" with different particle types, as it was in 1969 and again in 1972, concentrations varied widely over small ranges of altitudes and were often quite different on adjacent days. Averaging over a range of heights is therefore essential to avoid fortuitous bias from unusually rich (or poor) aerosol regions, and we have therefore arbitrarily divided the sulfate layer into "lower" (10-16 km), "middle" (16-22 km) and "upper" (22-28 km) regions for examining mean distributions and concentration trends.

### 3. Particle types

During 1968 and 1969 a much wider variety of particles was encountered than at any subsequent time except in January 1972. Some of these have been illustrated by Bigg and Ono (1971). During 1970 and 1971 simpler acid types were by far the most common, and a selection of these is shown in Fig. 1. All were collected on nitrocellulose strengthened with a thin film of carbon.

The appearance of the droplet rings depends to a large extent on the degree to which the collecting surface was hygroscopic and there is probably no real difference in the first three parts of Fig. 1 except for the proportion of non-acid material revealed by the central particle. However, the lower right photograph (which has been photographically reversed to appear like an optical photograph) shows quite clearly how a solid crystalline particle has left a crater in the nitrocellulose and carbon. On impact the central particle appears to have shattered, bounced and shrunk. The droplet rings show that it was mainly sulfuric acid. It is easy to see how larger particles of this type can be lost from an impactor. Figure 2 shows by way of contrast some of the particle types obtained during disturbed periods, also collected on nitrocellulose covered with carbon. Those on the top left show "splash rings", suggesting that they were completely liquid when impacted. Those on the lower left were the first evidence of the presence of new volcanic material in August 1971, and contain angular, electron-dense particles like those found by Mossop (1964). It is often impossible to distinguish and size the primary particles with certainty in such collections, and flights where they were present have been omitted from the analysis.

Figure 3 shows the appearance of particles such as those of Fig. 1 when collected on a copper surface. The etch marks and crystals consistent with those of copper sulfate suggest that the original particles were sulfuric acid.

#### 4. Size distributions as a function of altitude

Thirty-five flights in the five-year period May 1969 to April 1974 launched from Mildura, latitude  $34^{\circ}\text{S.}$ , longitude  $142^{\circ}\text{E.}$ , were selected for analysis. The only month not represented was December. In order to avoid the effects of stratification of the aerosol, cumulative size distributions have been arithmetically averaged over the altitude ranges 10-16 km, 16-22 km and 22-28 km, representing the lower, middle and upper regions of the stratospheric particle distributions. In Fig. 4 groups of curves, (a), (b) and (c), refer respectively to these three height ranges. Curve 1 in each group gives the overall mean for the 35 Australian flights while, for comparison, the remaining curves give previously published results reduced to a common scale. Curve 2 is the mean of the six Wyoming flights in 1972 (Bigg, 1975). Curve 3 is taken from Junge et al. (1961). Note that this curve in group (c) refers to "above 20 km" rather than the 22-28 km grouping that we have used. Curve 4 is taken from the "representative samples" given in their Table 2 by Ferry and Lem (1974) and refers to 1972-1973 impacted samples on 0.2 mm diameter palladium wire exposed from an aircraft. It was assumed in reducing their values to a common form with ours that their quoted  $10\text{ cm}^{-3}$  average concentration of particles larger than the detection limit of  $0.06\text{ }\mu\text{m}$  applied. Similarly the results of

Kondratyev et al. (1974) of particle counts collected on balloon-borne filters have been put in the same form by assuming a concentration for diameters  $>0.4 \mu\text{m}$  of  $0.3 \text{ cm}^{-3}$  to give curve 5. They used altitude groupings of 10-17 and 17-24 km. Curve 6 shows Mossop's (1965) 1963 distribution and curve 7 Friend's (1966) 1962-1963 distribution derived from the same type of impactor mounted on U-2 aircraft.

For particle diameters less than about  $0.5 \mu\text{m}$  it is clear that Mossop's (1965) and Friend's (1966) distributions are quite unlike the remainder. Cadle et al. (1973) have investigated the efficiency of this type of impactor and concluded that it collects only about 20% of the sulfate found by filters. Probably therefore the falling values for diameters less than  $0.5 \mu\text{m}$  represent decreasing collection efficiencies.

The relatively high concentrations of particles larger than  $1 \mu\text{m}$  diameter obtained by Kondratyev et al. (1974) using filters represents a marked departure from the present results, though they gave no details of "background" particles on the filters or whether these had been allowed for in the analysis.

The remaining results are in quite good agreement with one another (allowing for fluctuations of the aerosol content of the stratosphere) and probably give a fairly representative picture of the size distributions that occur. Note that none of the size distributions can reasonably be approximated by a simple power law. The "modified gamma distribution" proposed by Deirmendjian (1963) to represent stratospheric aerosol distributions,

$$n(r) = ar^{\alpha} \exp(-br^{\gamma}) ,$$

where  $n$  is the concentration of particles with radius  $r$  and

$a$ ,  $\alpha$ ,  $b$  and  $\gamma$  are constants, has often been used for model calculations. It produces the type of maximum and curvature increasing with diameter that is particularly evident in Fig 4, group (c). Values of  $\alpha = 2$  and  $\gamma = 0.7$  allow reasonable matching of the Australian data for the top level with suitable adjustment of the other constants, but the presence of slight inflections preclude an accurate representation by this formula.

#### 5. Seasonal variations in size distribution

When the Australian data is plotted by months it becomes evident that there is a significant seasonal variation at the lower levels that is relatively weaker in the upper levels. This is shown in Fig. 5 for January, April, July and October. These are the center months of three-month moving means (used to reduce the effects of longer-term changes). If instead the cumulative distributions are plotted by months, the nature of this seasonal change becomes more obvious. In the 10-16 km height range, shown in Fig. 6(a), an increased concentration of small particles reaches a maximum in mid-winter (July), the maximum occurring steadily later for larger particle diameters. This suggests an enhanced winter tropospheric-stratospheric air interchange providing reactive gases such as sulfur dioxide and ammonia with subsequent particle formation and growth.

At mid-levels (Fig. 6(b)) there is a slight winter maximum of particles in the middle size ranges and an enhancement of the small particles only in late spring (October-November). Finally, at the top of the particle layer (Fig. 6(c)) there is very little seasonal variation except possibly in the rare large particles. A very similar

result has been found by Hofmann et al. (1975), who showed that above 20 km the total aerosol concentration was almost constant whereas above 15 km a late winter maximum was evident. The curve emphasized in each figure is for diameters greater than  $0.23\text{ }\mu\text{m}$ , corresponding approximately to the range of particle sizes considered by Hofmann et al.

#### 6. Long-term trends

Since Fig. 6(c) shows that there is little seasonal variation in concentrations of particles larger than  $0.23\text{ }\mu\text{m}$  diameter in the 22-28 km height range this size and height range have been used in Fig. 7 to demonstrate longer-term changes. Because of its higher particle concentrations, the 16-22 km region is of more interest, so that trends for it are also shown. It must be recognized that part of the variation at this level may be seasonal. The gaps in this diagram are a result of the necessity of considering only group averages of several adjacent flights, for "patchiness" of the aerosol could lead to misleading information from single flights isolated in time.

The most noticeable feature of Fig. 7 is the very low concentrations (down by more than an order of magnitude) in 1971. The first indication of a subsequent increase was obtained with the isolated very large particles near the tropopause on 26 August, shown in the lower left of Fig. 2. Unfortunately the equipment failed at 15 km on that occasion. Weight restrictions prevented samples from being taken in October, but an automatic camera photographing the horizon at height intervals of 400 m showed on 5 October intense aerosol layers such as those that had been photographed in 1968 and 1969. In January 1972 large concentrations of particles of

very varied appearance were again found.

Volz (1974) has called attention to "the stratospheric dust event of October 1971" in the northern hemisphere, attributing it to an unknown volcanic event in the North Pacific. If this is a correct interpretation, the northern and southern hemisphere dust events must have been independent. Results from the January 1972 flights were not included in the time series because of frequent difficulties in identifying primary particles from the collections. Concentrations were probably higher than those shown for mid 1972 but reliable estimates were impossible.

Dyer (1974) has shown a short-lived dip in turbidity at Aspendale (latitude 38°S.) in mid 1971 and a sharp increase late in the year to a high maximum in early 1972. The generally poor agreement between Dyer's turbidity curve and our stratospheric particulate concentrations suggests that much of the turbidity is due to particles at the base of the stratosphere or in the troposphere. This points up the weakness in turbidity measurements if they do not have the benefit of additional information.

## 7. Conclusions

Because of the lengthy period that it covers and the large number of particles studied, the present work adds considerably to the meagre descriptions of stratospheric particle distributions that have previously been published. It shows quite close agreement with the pioneering work of Junge and his associates, who also sampled particles with an impactor carried on a balloon 10 years earlier. However, stratospheric particles generally appeared to contain a higher proportion of sulfuric acid than their illustrations

suggest. The seasonal variations in particle concentrations at 10-16 km and their absence at higher levels confirms and extends the results suggested by Hofmann et al. (1975).

Acknowledgments. The accurate measuring of photographs of tens of thousands of central particles, their shadows and their droplet rings was a task beyond our resources. It is with gratitude that we acknowledge the help provided by the Climatic Impact Assessment Program through the U.S. Office of Naval Research in funding assistants to perform this task. Those who contributed most to this work were Miss Mary Brauer, Mr Warwick Martin and Miss Ruth Hodge. I would also like to thank Mr W.J. Thompson, who maintained the equipment for the entire period of this experiment, and the staff of the Balloon Launching Station at Mildura for their help and cooperation over the five years.

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## FIGURE CAPTIONS

Figure 1 Typical stratospheric particles during undisturbed periods.

Top left: 25 June 1971, 23 km.

Top right: 3 May 1971, 26 km.

Lower left: 16 June 1971, 20 km.

Lower right: 25 April 1970, 20 km.

Figure 2 Stratospheric particles during disturbed periods.

Top left: 21 May 1969, 21 km.

Top right: 26 June 1969, 18 km.

Lower left: 28 August 1971, 12 km.

Lower right: 17 January 1972, 22 km.

Figure 3 Acid particles collected at 23 km on 13 March 1970 on a copper surface. The copper is etched and leaves crystals of copper sulfate.

Figure 4 The mean size distribution of aerosols.

(a) 10-16 km; (b) 16-22 km; (c) 22-28 km.

Curve 1 - the mean of 35 Australian flights 1969-1974.

" 2 - the mean of six Wyoming flights 1972 (Bigg 1975).

" 3 - Junge et al. (1961).

" 4 - Ferry and Lem (1974).

" 5 - Kondratyev et al. (1974).

" 6 - Mossop (1965).

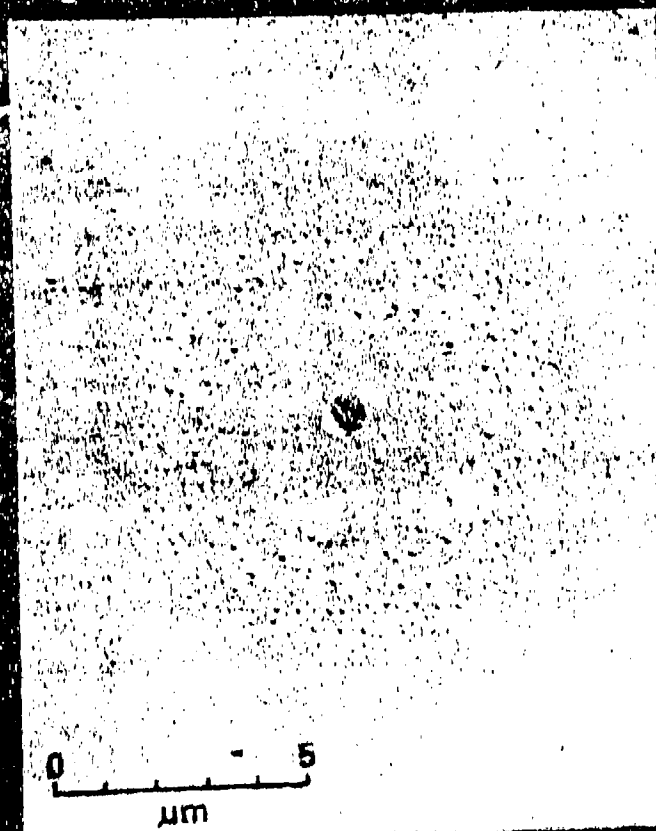
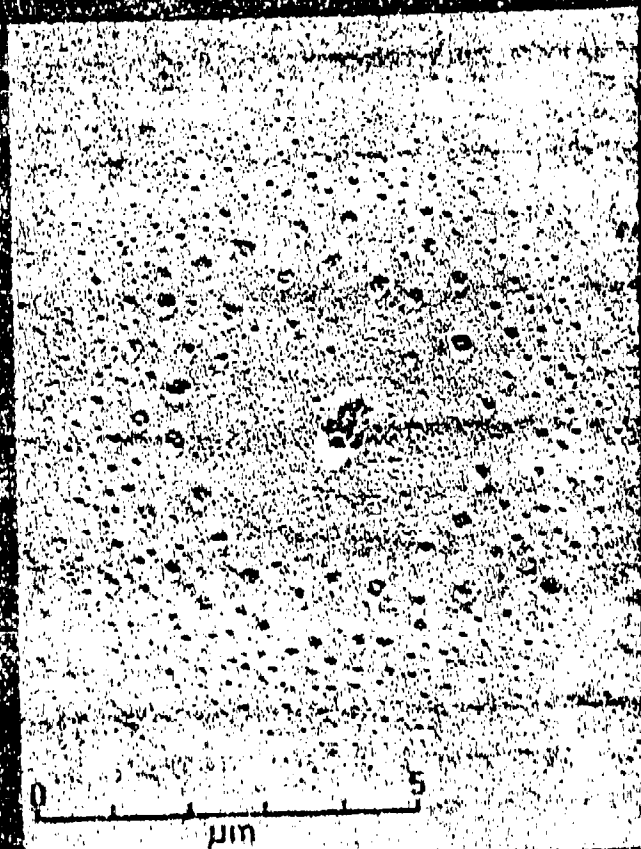
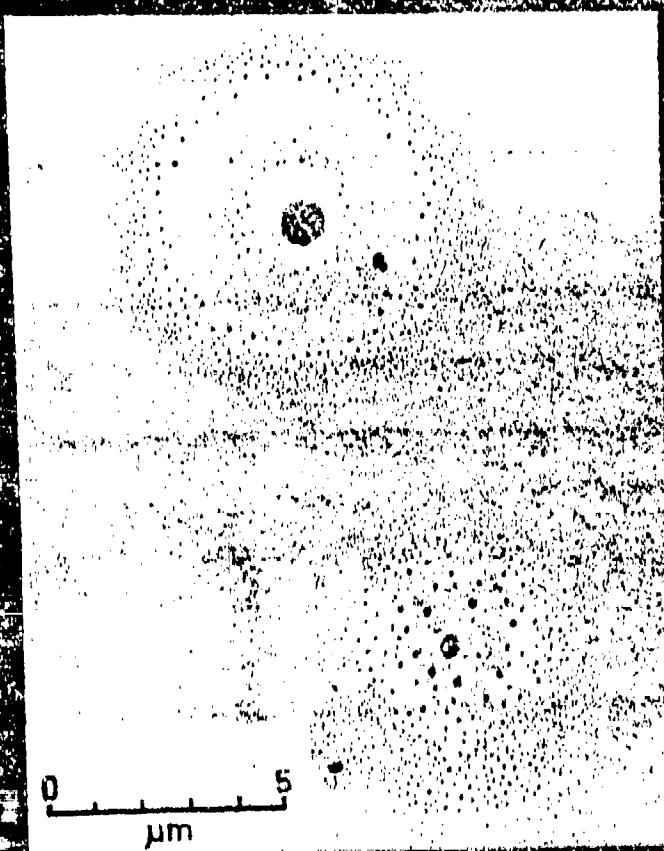
" 7 - Friend (1966).

Figure 5 Seasonal variations in size distributions.

(a) 10-16 km; (b) 16-22 km; (c) 22-28 km.

Figure 6 Seasonal variations in cumulative size distributions.  
(a) 10-16 km; (b) 16-22 km; (c) 22-28 km.

Figure 7 Trends in concentration of particles with diameters exceeding  $0.23 \mu\text{m}$ , 1969-1974.



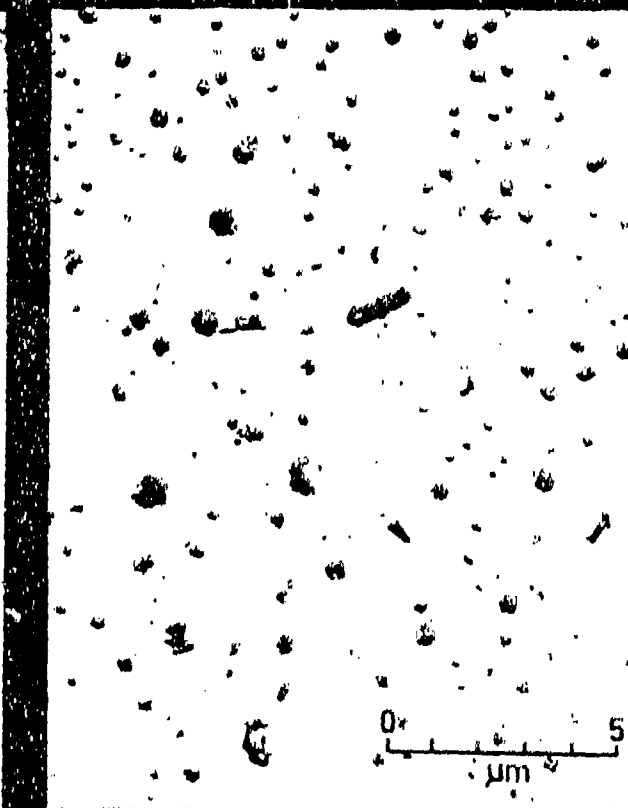
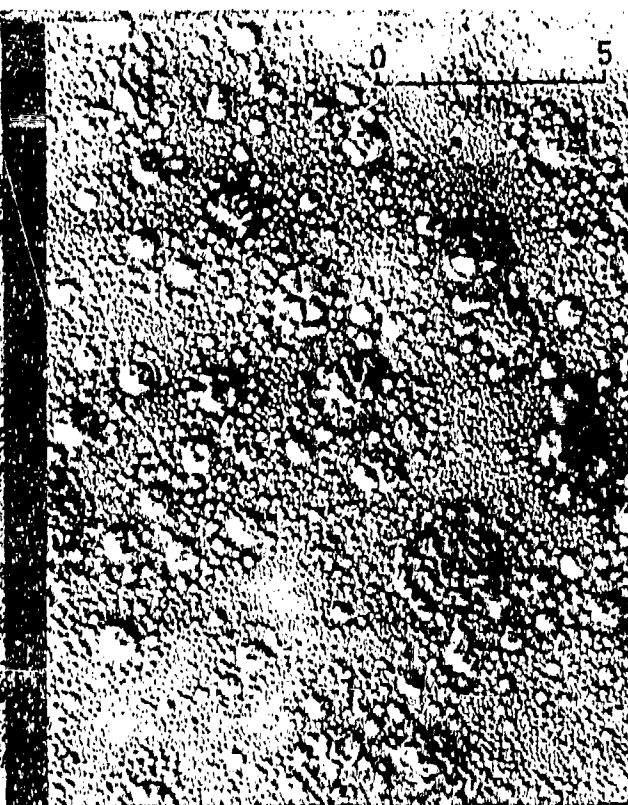
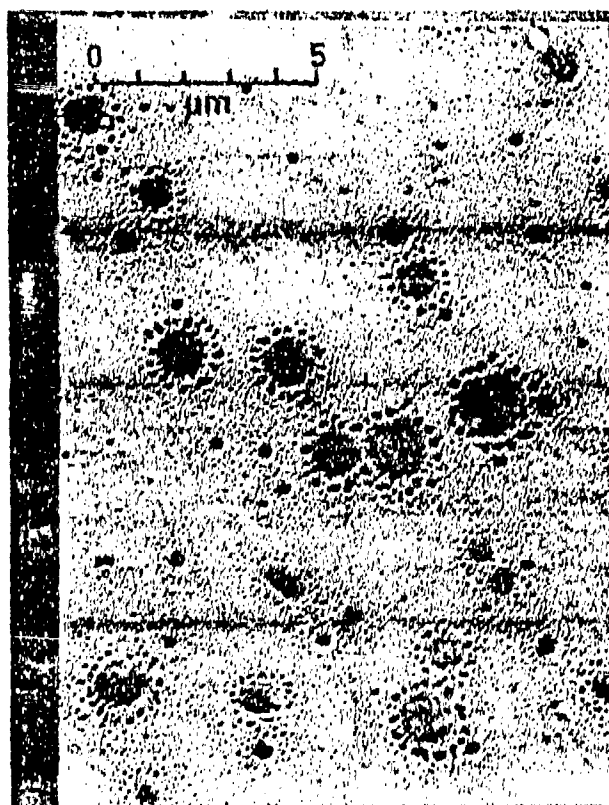
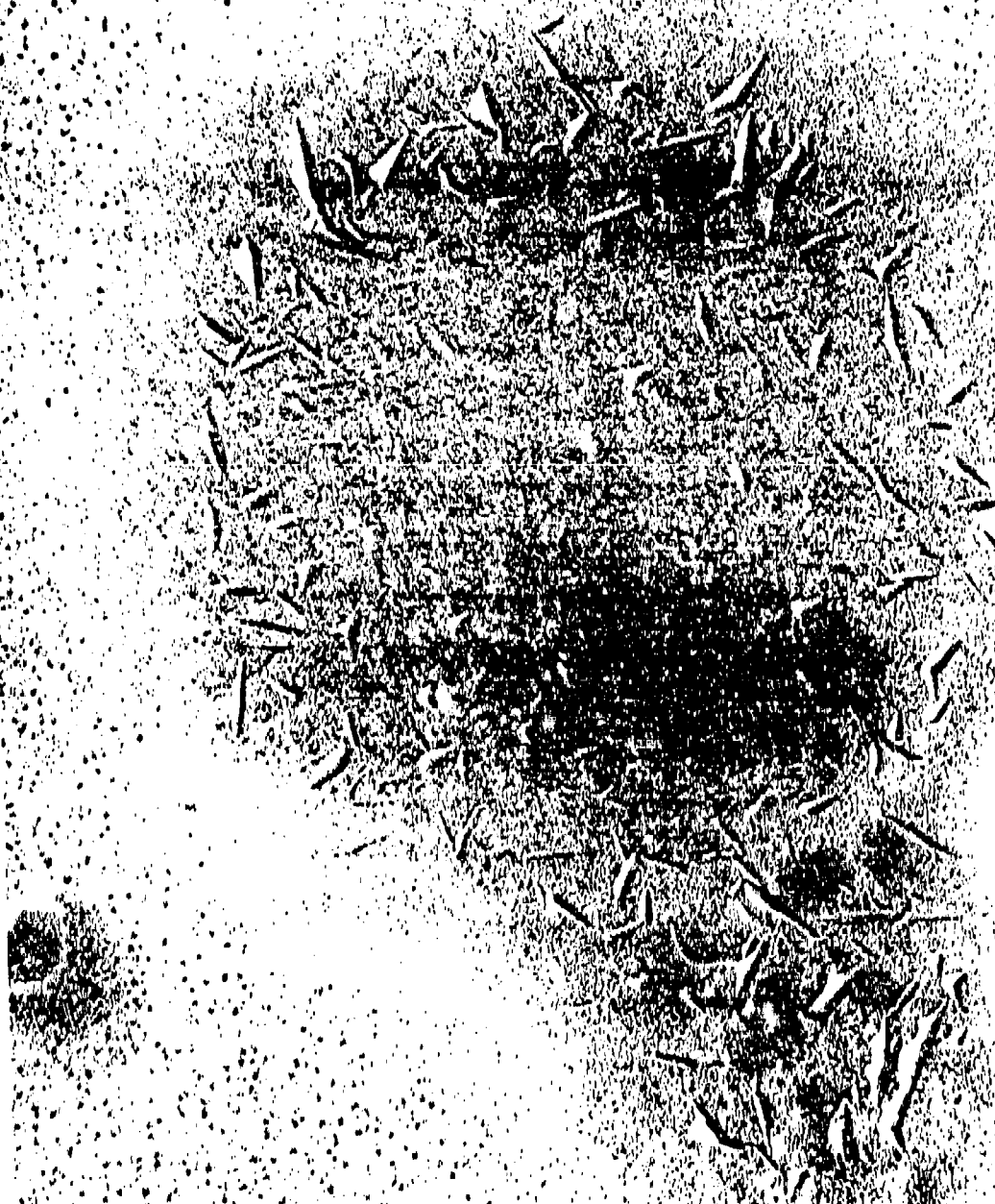


Fig. 2



1  $\mu$ m

Fig 3

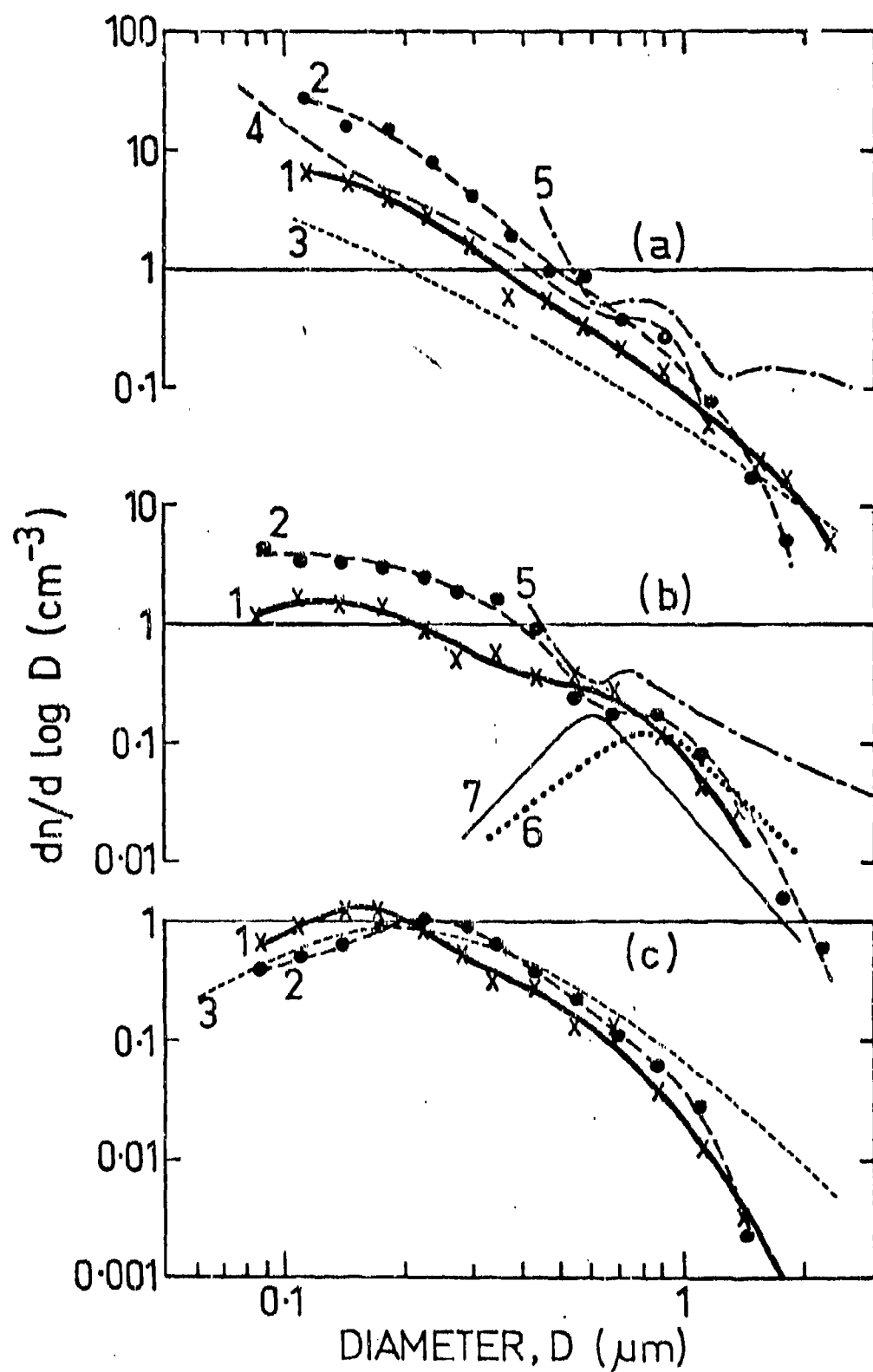


Fig. 4

