Electron microscope studies of noctilucent cloud particles

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ABSTRACT

Details of the sample preparation and handling techniques of the electron microscope samples for the noctilucent cloud sampling experiment are given. Typical electron micrographs and distribution functions are shown. Most of the noctilucent cloud particles appear to be extraterrestrial in origin. Approximately 10% of the total number of particles collected had a volatile coating which is believed to be ice.

1. Sample Preparation

A number of stringent requirements must be met in the preparation of substrates for the collection of small particles. The surfaces must consist of a film thin enough for easy penetration of an electron beam to permit their examination in an electron microscope. These films must be as free of contaminants as possible. Methods must be devised for the identification on the substrates of the inevitable contaminants other than by their physical appearance alone. The films must also be able to hold the particles and keep them free of any static charge that would cause particle repulsion during observation.

The means for supporting and handling such films to avoid contamination or loss of the collected particles is as important as the preparation of the substrates. Since the films had to be able to withstand considerable shock, acceleration, and vibration, they and their supports had to be firmly held in place in their sealed collection boxes. Control areas had to be provided since it is impossible to prepare electron microscopically clean films of the required areas. These control areas were obtained by shielding a portion of the collection films with an undercut section of the plastic hold-down clamps.

One of the cleanest types of substrate films that has been found suitable for electron microscopy is nitrocellulose. The nitrocellulose films used in this experiment were approximately 200 Å thick and cast from a 1 % amyl acetate solution on freshly distilled water. The amyl acetate solvent was refluxed over calcium oxide for four hours and then distilled in a one degree temperature range.

All cleaning and film preparation procedures were carried out in a clean box with air circulated through a thick filter. This box was located in a clean room the air of which was continuously circulated through Cambridge Absolute Filters. The 200 mesh electrolytic copper support screening, the screws, and other metal parts of the collection boxes were rinsed in clean acetone and blown dry with Freon gas dusters. The plastic clamps were wiped with benzene just prior to assembly. All parts were kept in sterile Petri dishes which were cleaned with Freon gas dusters. These Petri dishes were in turn kept in the clean box until the cleaning and assembly of the various components into the collection boxes was completed.

The screens were cut into wedge-shaped segments, cleaned and then dipped into a specimen screen adhesive solution and dried. The nitrocellulose film was cast on a freshly-distilled swept water surface and picked up on the screens. After drying, the films were shadowcast with aluminum in a vacuum evaporator. This process indexed initial contamination on the films for subsequent identification and provided a conducting surface for particle examination.

To determine the possible presence of a liquid or an ice that would become liquid before evaporation, a collection substrate was employed that was sensitive to both aqueous and non-aqueous liquids. This material was a red fuchsin dye which as a thin film would be deformed by droplets of liquid. This dye was deposited by a vacuum evaporation on nitrocellulose films prepared in the manner previously described.

In the event that a large particle should rupture the thin aluminum shadowcast and fuchsin coated nitrocellulose films and pass through, it would embed itself in a relatively thick film of methyl cellulose. Before the methyl cellulose films were mounted they were given a quick dip in acetone, a good solvent for methyl cellulose. This process cleaned the methyl cellulose and also produced a chemically polished surface thus eliminating surface marks.

Since no significant results were obtained with the indium surfaces, details of their preparation and handling have been omitted. The preparation and study of the calcium film moisture indicators are described in Ref. D.

To facilitate handling of the screens during the loading and unloading of the collection boxes, the covered screens were cemented to identical wedge-shaped pieces of plastic 1.6 mm thick. Each of the four different experimental surfaces in each collection box had the same size and shape and could be placed in any position on the aluminum supporting plate. The ring-shaped plastic hold-down clamps were milled out in certain areas between the screw holes so that the plastic did not come in contact with corresponding areas of the film-coated screens. These shielded areas constituted the control areas previously mentioned.

All tweezers, needles, and scalpels used for manipulating and picking up various components of the particle collectors were cleaned by wiping, then forcing the points through lens tissue and tearing out. Hands were washed with soap and water immediately prior to component manipulation operations. Clean uncovered hands generate only skin tissue particles which have a very characteristic structure and are easily recognized in an electron microscope.

To insure that collection films having the utmost cleanliness were used, all mounted films were prepared in duplicate under identical conditions of material manipulation, shadow casting and so forth. Immediately before mounting of a film-coated screen on the plastic support plate, a specimen screen was punched from one of the two samples chosen randomly and checked for cleanliness in an electron microscope. If any undue contamination or unidentifiable material was found, another screen that had been checked was used. The tested screen duplicates of the screens used were retained in clean boxes for use as laboratory control samples for future study if needed.

The assembled wedge-shaped components were fastened to the aluminum supporting plates with screws and the plastic hold-down clamps. The supporting plates were then screwed firmly to the insides of the cleaned collection boxes. The sealing covers were put on the collection boxes and the sealed boxes placed in plastic bags. An open collection box loaded with the four different experimental surfaces along with a collection box cover is shown in Fig. 6 of Ref. A.

2. Sample Handling

To permit loading of the sample boxes into the rocket nose cones under clean conditions, a portable glove box facility was constructed and carried to Kronogård, Sweden. This box was kept under positive pressure with air filtered through Cambridge Absolute Filters. Prior to loading the sample boxes into the rocket nose cones, an extensive effort was made to clean the inside of the glove box, the rocket payloads, the sample boxes, the gloves and all needed tools. This cleaning which took several days, included washing the inside of the glove box with acetone and ethanol, wiping with clean lint-free nylon rags and finally vacuum cleaning.

After the nose cones were loaded with the sample boxes the nose cones were each covered with a clean sealed plastic bag. During the initial acceleration of the rocket, these plastic bags were ripped off by tie lines held to the rocket launcher. The entire payloads, telemetry and recovery sections were enclosed in an additional plastic bag through which heated dry, filtered clean air was passed during the waiting periods between possible flight times.

After the first flight, which was flown in the absence of noctilucent clouds, the sealed boxes (numbers 5 and 9) were removed from the rocket nose cone and the exposed edges between the covers and sample boxes were covered with plastic cement to provide additional protection against air leaks during storage. The nose cone of the second flight, which was flown through a noctilucent cloud display, was damaged opon ground impact and could not be opened in Sweden. This nose cone was returned to the clean laboratory where it was carefully cut apart in a lathe and boxes 11 and 14 safely recovered. These boxes as well as boxes 5 and 9 were found to be properly sealed. No useful materials were recovered from the third and fourth flights.

After the flights, the gases in the boxes 5, 9, 11, and 14 were removed for mass spectrographic analysis by inserting a hypodermic needle into a silastic filled hole at the base of the boxes. After removal of the gases, the boxes were returned to the clean room in which they had been prepared. The boxes were brought to atmospheric pressure with dry filtered air from a large hypodermic syringe inserted through the silastic filled hole in the bottom of the boxes. When the internal pressure reached ambient the covers were removed. As before, extreme precautions were taken to prevent airborne contamination. Also, all tools and storage boxes were carefully wiped clean with benzene on lens paper or dusted with Freon gas.

After removing the aluminum supporting discs holding the collecting films from the boxes, all elements were disassembled by holding the boxes and components in such an altitude that no chips or other possible contaminants from the removal of the screws would fall on the films.

The film-covered screens with their attached methyl cellulose film backing were separated from the other support elements and placed in individual clean plastic boxes. A cleaned electron microscope specimen screen punch was used to punch out specimen screens with the film backing of both the exposed area as well as unexposed control areas under the hold-down clamp ring. Each screen with its backing film was placed in a gelatin capsule. Each gelatin capsule was then placed in a clean labelled plastic box for storage until future use.

3. Electron Microscope Results

A preliminary examination of the flight control surfaces revealed that the cleanliness of the surfaces had indeed been maintained during the many operations carried out with them. It was found that the particle densities on the samples which had passed through the noctilucent cloud were approximately three orders of magnitude higher than the particle densities on the various control surfaces. During the preliminary scanning of the aluminum

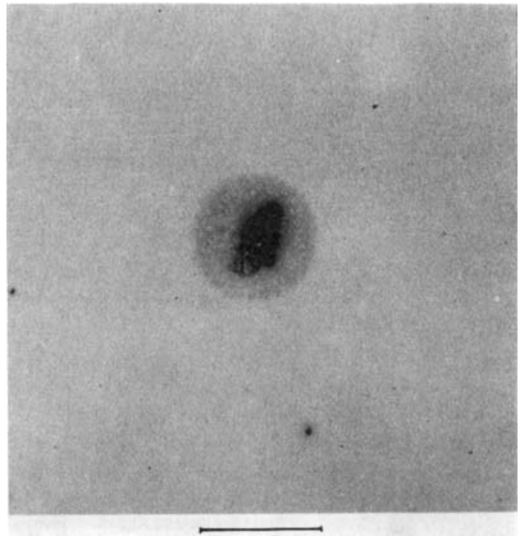

FIG. 1. Electron micrograph of particle collected in the noctilucent cloud flight on an aluminum-shadowed nitrocellulose sampling surface. Scale = 1 micron.

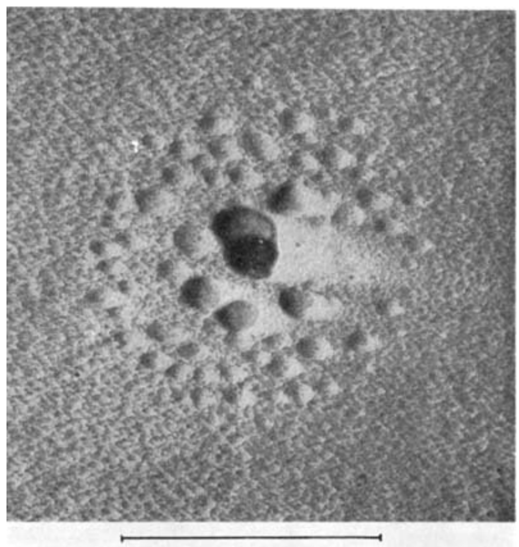

FIG. 2. Particle similar to that in Figure 1 but with additional Cr shadowing at near-grazing incidence ($\approx 5\%$). Scale = 1 micron.

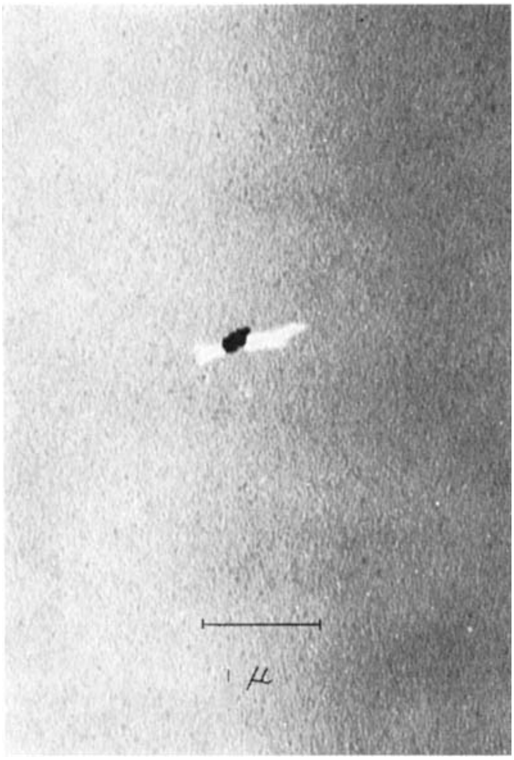

FIG. 3. Electron micrograph of double-shadowed contaminant particle.

shadowed nitrocellulose samples, it appeared that some of the particles had poorly-defined structures around them. Fig. 1 shows an electron micrograph of this type of particle. To bring out the details of these structures, it was decided to apply an additional shadowing with chromium at near-grazing incidence (5°). Fig. 2 shows a particle similar to that shown in Fig. 1 after the second shadowing. It will be noticed that the particle has a symmetrical pattern or halo around it. This halo has a number of symmetrically-placed humps in it. It appears that this pattern is the result of the interaction of a volatile material with the nitrocellulose film. We believe that this volatile material was ice which surrounded the particle prior to its collection.

The second shadowing also provided an additional criteria for discrimination against contaminant materials which were always present. For if a particle had two shadows it must have been deposited before the first shadowing or if a particle had no shadows it must have been deposited after the second shadowing or have been on the underside of the film or possibly within the tin film. Fig. 3 shows a double shadowed contaminant particle.

Most of the samples collected were then given the second shadowing. The samples were then equally divided for study by three independent teams of investigators. The following

TABLE 2. *Noctilucent cloud particle study.*

Sample Type	Box Nr.	Total Nr. of Part./(Meter)^2
Nitrocellulose	14	7.6×10^{10}
Nitrocellulose	11	1.9×10^{10}
Fuchsin	14	4.5×10^{10}
Fuchsin	11	2.4×10^{9}
Fuchsin	11	4.7×10^{9}
Fuchsin	14	1.9×10^{10}
Nitrocellulose	11	2.1×10^{10}
Fuchsin	14	8.3×10^{10}
Nitrocellulose	14	4.3×10^{10}
Fuchsin	14	2.5×10^{10}
Nitrocellulose	14	3.4×10^{10}

TABLE 1. *Control study.*

Sample Type	Box Nr	Contaminant Particles	Extraterrestrial Particles	Area Scanned (mm)^2	Total Part./(Meter)^2
Nitrocellulose	9	8	18	5.61×10^{-1}	4.3×10^{7}
Nitrocellulose	9	6	6	1.2	1.2×10^{7}
Fuchsin (Control)	9	3	—	7.73×10^{-1}	3.9×10^{6}
Fuchsin (Control)	9	9	—	7.30×10^{-1}	6.9×10^{6}
Nitrocellulose (Control)	9	11	—	6.76×10^{-1}	1.6×10^{7}
Nitrocellulose (Control)	5	30	—	7.85×10^{-1}	3.9×10^{7}
Nitrocellulose (Control)	5	15	—	8.35×10^{-1}	1.8×10^{7}
Nitrocellulose	5	17	4	8.35×10^{-1}	2.5×10^{7}

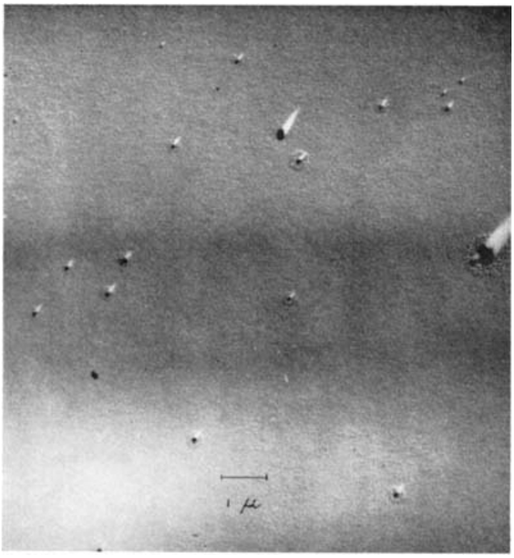

FIG. 4. Electron micrograph of portion of a nitrocellulose sampling surface from the noctilucent cloud flight. The picture shows a typical field of view. The halo structure surrounding a number of the particles should be noticed.

data are representative of the work of all groups, each of whom utilized slightly different techniques for their systematic scanning of the nitrocellulose and fuchsin surfaces.

Table 1 shows typical numbers of particles per unit area obtained from study of the control and experimental surfaces of boxes 5 and 9. It will be seen that the contaminant and extra-

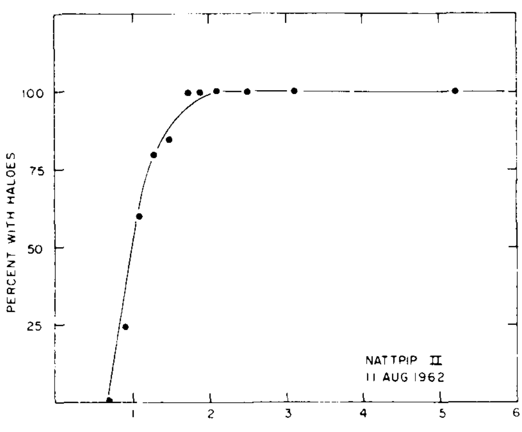

FIG. 5. Percentage of particles having the halo structure as function of the average diameter of the solid nucleus. The curve refers to the noctilucent cloud flight.

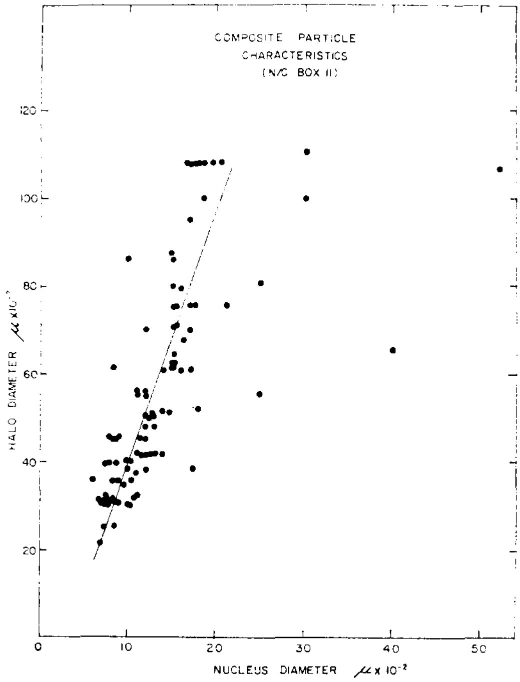

FIG. 6. Scattergram of halo diameter versus the diameter of the nucleus. The curve refers to the noctilucent cloud flight.

terrestrial particle densities on these substrates are of the order of 2×10^{7} particles per square meter. Table 2 shows equivalent particle densities obtained from the study of the materials of boxes 11 and 14 which sampled the noctilucent cloud. It will be noted that the particle densities in Table 2 are generally in the range from 2×10^{10} to 8×10^{10} particles/meter^2. The

TABLE 3. *Fraction of total number of particles with halos.*

Sample Type	Box Nr.	Per-cent Particles With Halos
Fuchsin	11	15.
Fuchsin	11	3.7
Nitrocellulose	11	44.
Nitrocellulose	11	20.
Fuchsin	14	7.5
Fuchsin	14	6.5
Fuchsin	14	4.8
Fuchsin	14	17.
Nitrocellulose	14	13.

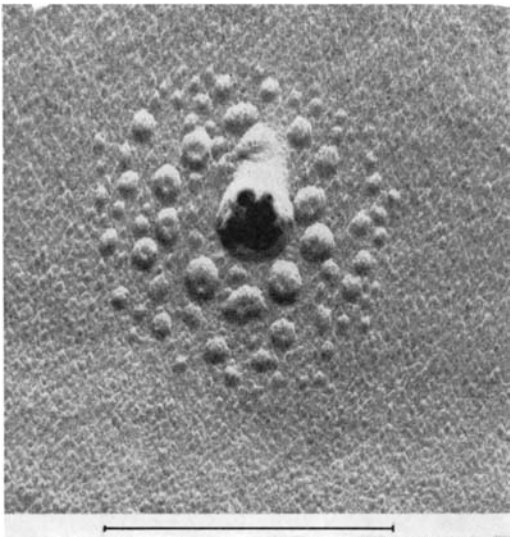

FIG. 7. Noctilucent cloud particle with halo and composite nucleus with dark circular-shaped inclusions. Scale = 1 micron.

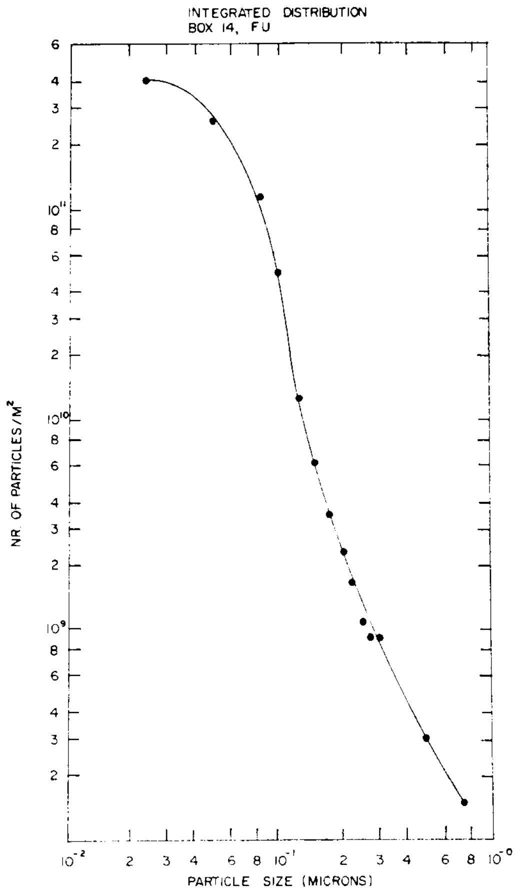

FIG. 9. Integrated size distribution of noctilucent cloud particles collected on the fuchsin-coated sample in box 14. The curve shows the total number of particles larger than a given size per unit surface. The points were obtained by measuring particle sizes from a large number of intermediate magnification electron micrographs.

particle density in box 14 was approximately four times as great as that in box 11. The particle density also varied somewhat as a function of position in a given box. Fig. 4 shows a typical field of view showing a number of noctilucent cloud particles, some of which have the halo structure mentioned above. Fig. 5 shows how the percentage of particles having the haloed structure varies with the size of the particle nucleus. It will be seen from Fig. 5 that almost all of the noctilucent cloud particles having nuclei larger than 0.2 micron have the haloed structure whereas most of the particles smaller than 0.1 micron do not. From Table 3 it can be seen that the total number of noctilucent cloud particles having visible halo structures generally falls in the range from 5 to 20 %. Fig. 6 is a typical scattergram showing the relationship between the halo diameter and the diameter of

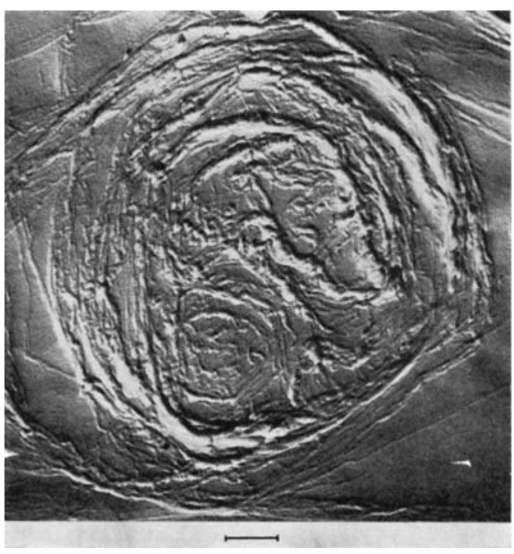

FIG. 8. Structure found on the nitrocellulose film which appears to be result of interaction of the substrate with volatile particle which subsequently evaporated. Scale = 1 micron.

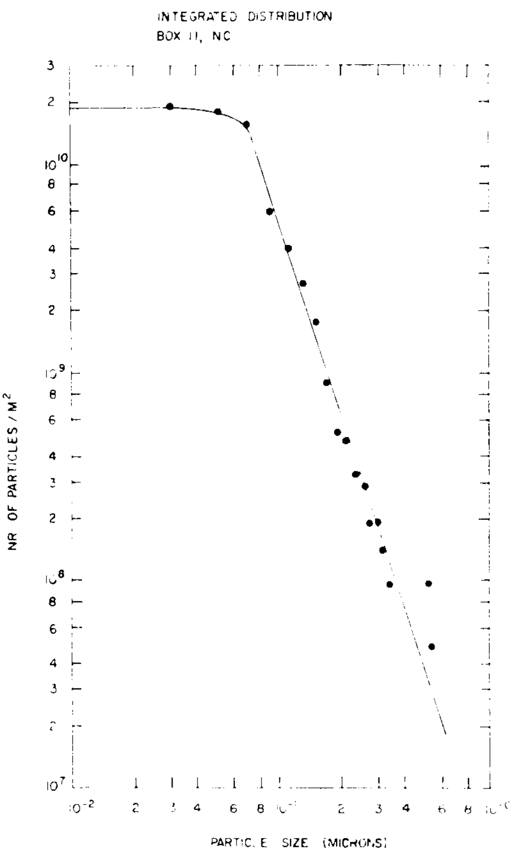

FIG. 10. Integrated size distribution of noctilucent particles from the nitrocellulose sample in box 11.

the nucleus of the haloed-type noctilucent cloud particles.

Fig. 7 shows a haloed noctilucent cloud particle with a composite nucleus which appears to contain several spherical inclusions. Such spheres in nuclei have been found quite frequently, although not usually in the artistic arrangement in which they appear in Fig. 7.

The noctilucent cloud particles and nuclei are similar in appearance to smallest particles found in the Venus-Fly Trap experiment (HEMENWAY and SOBERMAN, 1962) and other high altitude collections (HEMENWAY, FULLAM and PHILLIPS, 1961). Fig. 8 shows a structure in a nitrocellulose film which appears to be the result of an interaction of the nitrocellulose substrate with a volatile particle which subsequently evaporated. Several dozen such structures have been found in the scanning of the noctilucent cloud samples, but none on the controls or control flight.

Fig. 9 shows an integrated size distribution (ie, the total number of particles larger than a given size) of noctilucent cloud particles obtained by measuring particle size from electron micrographs of a fuchsin-coated sample from box 14. Fig. 10 is an integrated size distribution from the nitrocellulose sample from box 11. The scanning technique used for the data for Fig. 10 was to use a sufficiently high magnification that on the average about one third of a particle was seen per field of view. Then, pictures were taken of all particles individually and the total area scanned was measured. It will be noted that the general appearance of the curves is about the same as that in Fig. 9 except that the size cutoff appears to be somewhat sharper and the slope somewhat more constant. Very few noctilucent cloud particles were found smaller than 500 Å in diameter although 100 Å particles should have been readily detected by these techniques.

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