Electron microscope photographs of extraterrestrial particles

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ABSTRACT

With suitable collection methods it is possible to distinguish particles resident in the stratosphere from contaminants and to identify those of extraterrestrial origin. A catalogue of physical appearances under an electron microscope is presented which shows the most common types of such particles encountered in about 40 collections made from balloons in the course of three years. We conclude that small extraterrestrial particles are most commonly fragile low-density assemblages of still smaller particles, frequently composed of crystalline material and often associated with liquid or solid volatile components. It is suggested that these are of cometary origin.

Introduction

Discussing the results of particle collections from high-altitude rocket flights, Farlow (1968) concluded: "... it must be realized that little, if any, reliable information now exists to describe the physical and chemical characteristics of micrometeoroids near the earth". The situation has not changed greatly since then, although Hemenway & Hallgren (1969) have described one particle type apparently collected in flight (Farlow considered this to be a probable contaminant), while Farlow, Ferry & Blanchard (1960) have described two others.

A catalogue of physical appearances under an electron microscope of all particles having a reasonable chance of being extraterrestrial would greatly reduce the difficulty of recognition. From such a catalogue it would be possible with any new collection of particles to select the most likely candidates for more detailed analysis.

At first sight it seems highly improbable that a useful list of particle types could be produced, for an infinite variety might be expected. It also seems unlikely that any simple collection system from a balloon could allow the identification of cosmic material with any certainty. It is our aim to show that the 30 to 40 km altitude region is a particularly favourable one for sampling extraterrestrial particles and that a few distinctive types account for a large proportion of them. Their concentration is relatively high below 40 km because their fall velocities are low, the region is thermally very stable and any mixing from lower levels is readily detectable by the very characteristic particles associated with it.

Collection and examination procedures

Particles were collected from high-altitude balloons launched from Mildura (latitude 34° S) or Longreach (latitude 23° S) in Australia. The balloons ascended at about 250 m min^{-1} and were maintained at a float altitude (the highest altitude 40 km) for a period of one to four hours or more. Recovery after descent was usually accomplished with little delay. The specimens were shadowed and examined at the Electron Microscope Unit at the University of Sydney.

At first we used a simple impaction system at the float altitude only. Electron microscope screens using carbon-coated nitrocellulose films as the collecting surfaces were placed 1 mm below 1 mm diameter holes through which air was sucked by a vacuum pump. Flow rates were about 10 l min^{-1} to an altitude of 20 km and fell off to about 3 l min^{-1} at 36 km. The system had the great advantage that particles were collected on less than one-tenth of the area of each screen, allowing most of it to be used as a control for contamination arising during handling. Large particles were removed in a sedimentation chamber to prevent them from damaging the film at the high impaction velocities used. This also protected the screens

from contamination during launching and landing of the payload. The results have been described by Bigg, Ono & Thompson (1970).

The next improvement was to include a similar system for use during ascent, in which the electron microscope screens were arranged in a circle and made to rotate slowly beneath the hole. This allowed changes in particle types, sizes and concentration with altitude to be recorded with a height resolution of about 100 m above 15 km altitude.

Another collection system was used to detect the large particles excluded by the impactor. This consisted simply of two thin arms 15 cm long, rotated at 3 000 r.p.m. by a small motor. Electron microscope screens in small tubes faced forward for collection and backwards for controls. A pressure switch turned the motor on and later off at about 30 km altitude. The screens were unprotected during ascent and until recovery and we had expected severe contamination problems. In fact, the control surfaces have been so clean that identification of particles collected in flight would have been easy even without reference to the simultaneously obtained specimens from the impactors. In a later design, the screens lay behind conical holes, which protected them and concentrated the particles on to a smaller area.

Upon recovery the electron microscope screens were shadowed in a vacuum at a 30° angle with a gold-palladium alloy before being examined in an electron microscope. The shadow increased contrast and its length allowed the height of the particle to be estimated. Farlow (1968) remarked that shadowing with gold leads to the production of many small spheres. This does not appear to have been a difficulty with the gold-palladium alloy, as small spheres were quite rare and usually accompanied by semi-liquid material obviously not due to shadowing. In the usual photographic representation shadows are dark while the electron-dense areas are black. In the figures included here electron-dense areas are white and electron transparent areas are dark in order to resemble optical photographs.

Particles of the upper troposphere and lower stratosphere

It is relatively easy to establish that the particles collected by impaction through a small hole were obtained during sampling, for most of them were concentrated in a spot less than one-tenth of the area of the electron microscope screen. (The only exceptions were long rigid chains of particles having a surface contact area very small compared with their total surface area. These naturally tended to migrate outwards towards the edges of the screen in the horizontally directed blast of air.) The particles on the remainder of the screen should be representative of those acquired during handling.

The outer parts of the screen, or any part of the control screens which have been included in the package but not exposed, usually contain about two particles larger than 0.1 μm diameter per grid square of 100×100 μm compared with 100 to 10 000 in the centre of the impaction line. This represents the degree of contamination arising during preparation and handling. By the standards necessary in rocket experiments this is not very good, but because of the overwhelmingly greater number of particles collected in flight it is perfectly adequate in our experiments. By restricting our discussion only to common types the possibility of including them is removed.

Establishing that the very numerous particles within the impacted part of the screens were not shed from the balloon, its associated equipment, or the impactor itself relies on the following considerations:

(1) Such particles would be present at all altitudes. Instead we find that particles of different types and sizes are usually specific to rather narrow altitude ranges.

(2) The great majority of particles below 25 km are liquid or semi-liquid on collection, leading to a highly flattened appearance. It would be virtually impossible to dislodge such particles less than a few microns in diameter from a surface.

(3) Terrestrial dusts of the dry inland where these flights were made do not react with thin films of aluminium or copper. Most of those collected at altitude react with aluminium and all of them react with copper. These tests will be reported in detail in a later publication.

On two flights only, small polyethylene spheres of the type used to lubricate the folds of the balloon, were collected. These of course are completely different from the particle types

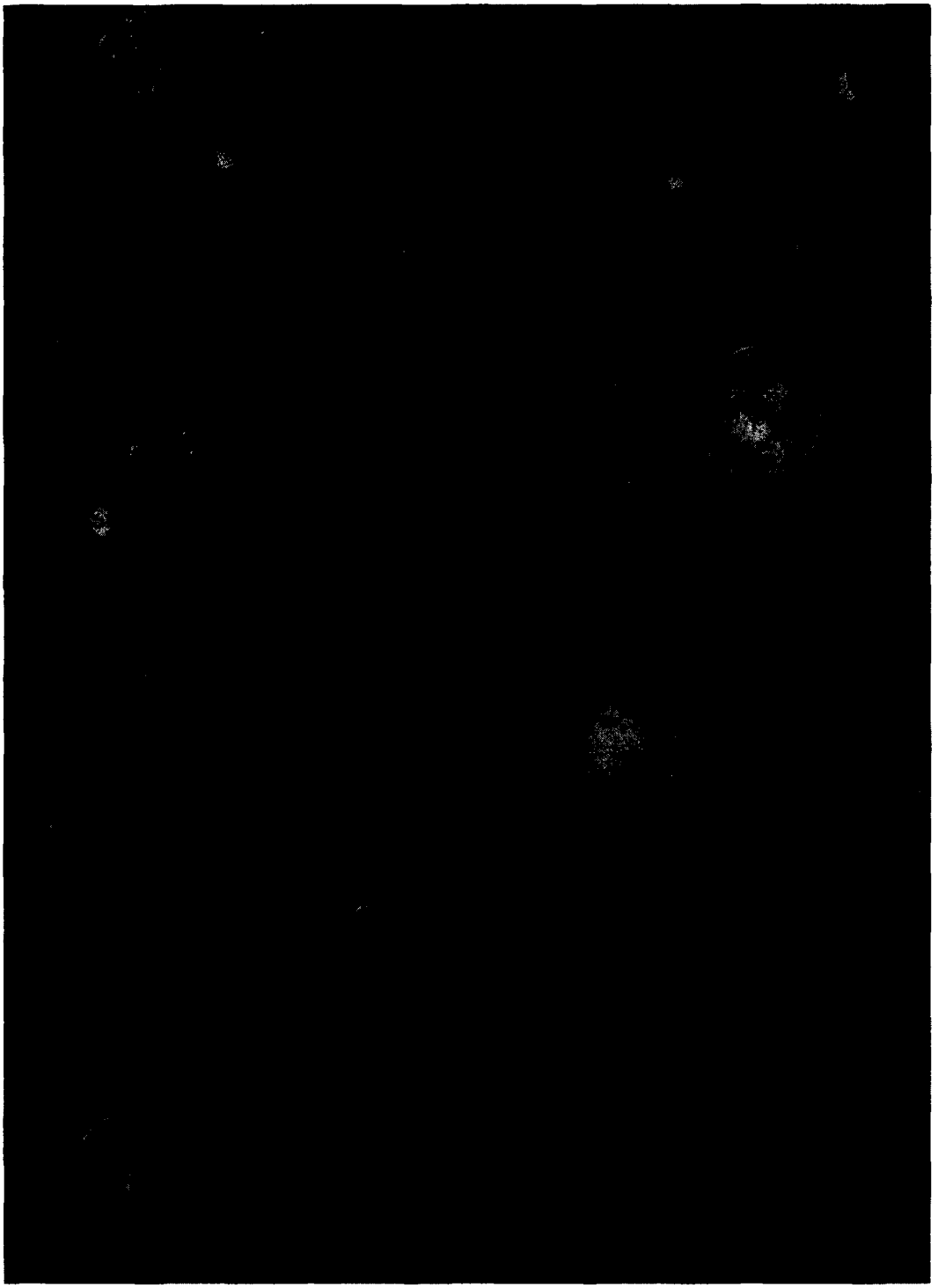

Fig. 1. Typical group of atmospheric particles collected at a height of 5 km.

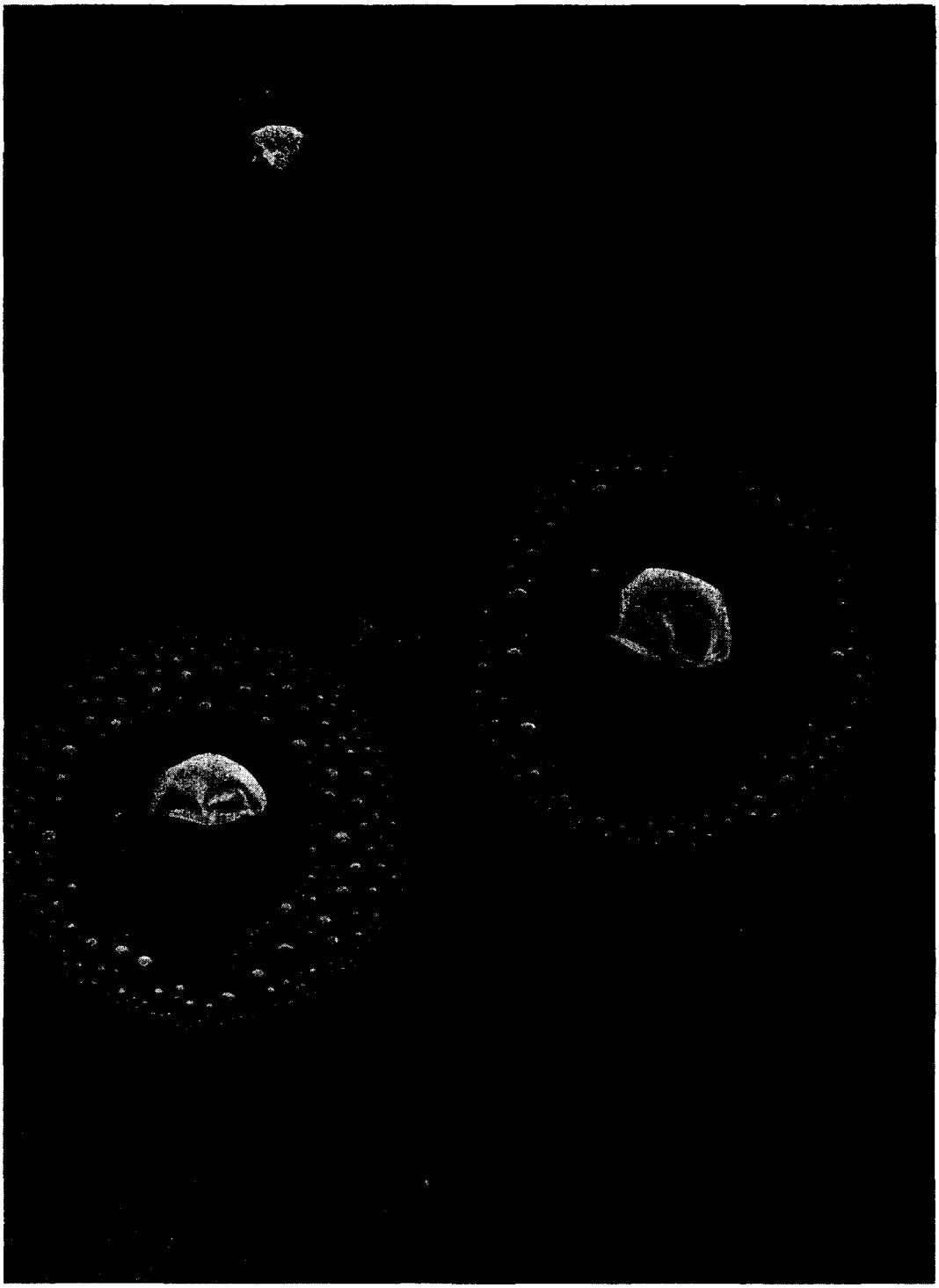

Fig. 2. Particles with a high sulphuric acid content, typical of the 15–25 km region.

to be described. Their presence is, however, a useful guide to the possibility of contamination by other particles from the balloon.

In the mid-troposphere in the absence of dust storms the most common particles encountered are liquid or semi-liquid types which spread out almost flat on the screen. Solid electron-dense particles also occur but are usually in the minority. The situation would perhaps be different in more populated parts of the globe. The photograph of Fig. 1 of a collection made at 5 000 m shows a typical group, most of which can be duplicated in the laboratory by evaporating ammonium sulphate, bisulphate or persulphate solutions at reduced pressure and impacting the particles formed on to an electron microscope screen.

In the lower stratosphere the size distribution, concentration and physical appearance of the particles may change very greatly within vertical distances of only 100 m and several distinct layers are usually found between 15 and 25 km. Most of the particles within this region have however one feature in common —a high sulphuric acid content. This is revealed by symmetrical rings of satellite droplets around a central particle, such as those shown in Fig. 2. For a fuller description of these particles see Bigg, Ono & Thompson (1970).

It is interesting that before the eruption of the volcano Mt. Agung in 1963, Mossop (1965) found no free acid but only ammonium sulphate particles and Friend (1966) ammonium sulphate and persulphate at around 20 km altitude. Junge & Manson (1961), however, showed some particles from the same altitude which appear to have had some free acid. Either volcanic emission of sulphur gases has been greater since 1963 than before it, or it has taken a very long time to restore a sufficient concentration of ammonia to neutralize the excess acid consequent upon the emission of sulphur dioxide in 1963.

Above 25 km the sulphuric acid drops become rarer, though isolated layers of low concentrations have occurred at higher levels. We have used the presence of such particles as indicators of the possibility of upward air interchanges. Because such interchanges might bring with them particles of terrestrial origin, less emphasis is placed on collections from altitudes where they occur. Skrivanek (1967) has demonstrated the presence of sulphuric acid in particles thought to have been collected from rockets in noctilucent clouds near 80 km, so that their presence at 35 km is not *necessarily* an indication of upward air movements.

Particles above 30 km

Above about 30 km almost none of the particles has free sulphuric acid but many are accompanied by a volatile coating of an entirely different sort. It apparently spreads over the screen typically to a diameter of 10 μm around a 1 μm diam. particle, and often crystallizes in a hexagonal form. During storage or shadowing of the screen it evaporates, sometimes almost completely but interferes with the deposition of the gold-palladium atoms and so leaves a visible stain.

The presence of these stains ensures that the particles were resident at high altitude when collected, for they have not been found in the troposphere, nor on particles showing free sulphuric acid.

There are two types of liquid stains which have been occasionally observed on particles of the lower stratosphere or troposphere but because they are much more common at high altitude are considered helpful in identifying the extraterrestrial types. In one of these the liquid forms a smooth symmetrical ring around the particle, casting a short shadow. (Examples may be seen in Figs. 4 and 7). The other type of liquid appears to wet completely the surface near the particle so that it leaves no shadow. The liquid increases gradually in thickness towards the particle; being quite electron-dense it often obscures the centre of the parent body.

In the past we have considered dry particles to have been collected at high altitudes if very similar types showing rings or stains were collected simultaneously. A more positive test which we have recently developed is to substitute copper or aluminium for the carbon coating on the nitrocellulose film. It seems that particles in high-altitude samples which appear dry on carbon films often etch aluminium and nearly always show strong reactions with copper, suggesting the presence of reactive but highly volatile liquids.

One further possibility must be considered. The chemicals which evaporate from incoming meteors mainly above 90 km will diffuse down-

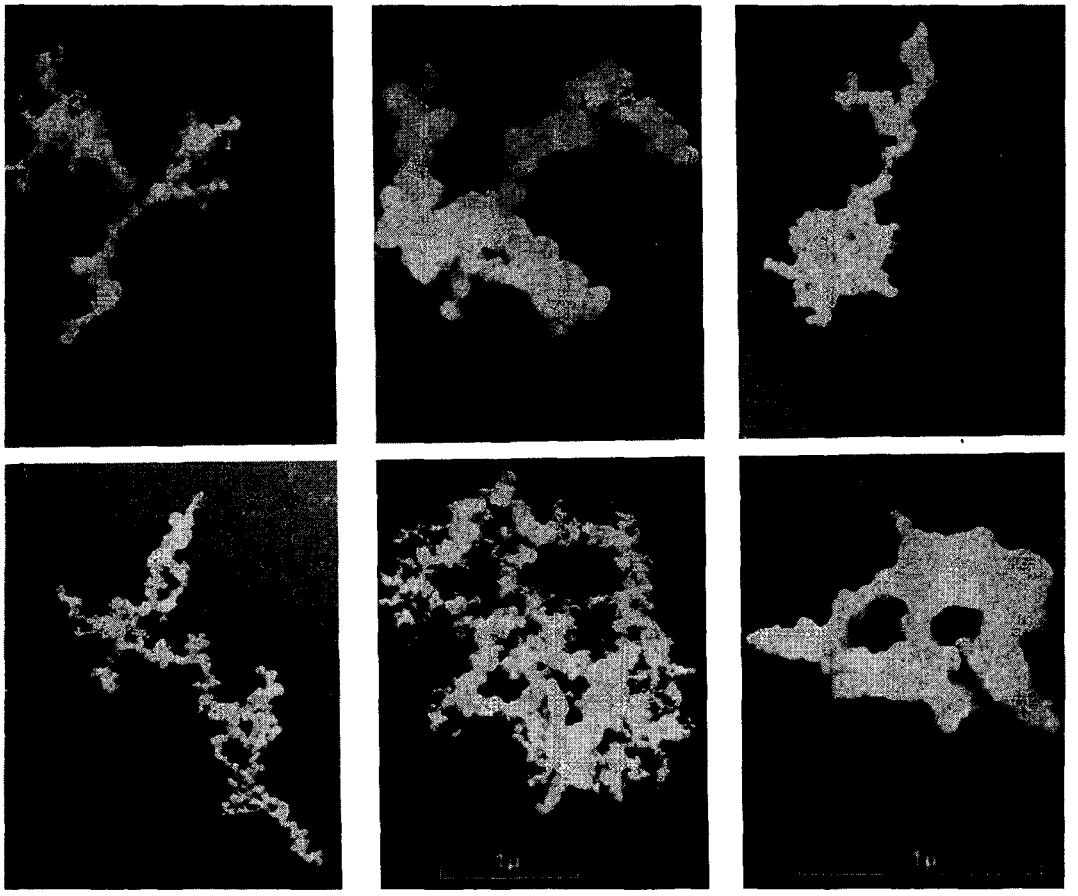

Fig. 3. Intricate chains of very small particles—obtained from attitudes above 25 km.

wards to the temperature minimum at the mesopause. If they are present in sufficient quantities, the creation of a supersaturated vapour is possible so that the small solid fragments falling through the region may become coated with recondensed products. In addition to this, photochemical reactions of the gaseous constituents are also likely. Thus some of the incoming particles may have had their appearance or composition modified by passage through the atmosphere. This is least likely for the larger fragments which will have had a very short residence time near 80 km altitude.

We therefore feel that it is reasonable to identify as meteoric those particles in the impaction spot which have no counterparts on unexposed screens, or very few screens exposed at lower altitudes, and describe them in more detail below. It should be noted however that the appearance of the particles must depend on the nature of the collecting surface because of the volatile or reactive components. In all cases illustrated here a carbon coating on a nitrocellulose base was used.

Electron microscope photographs of extraterrestrial particles

While many of the particles obtained are superficially very different, and the groups represented in the sample on one day may be quite distinct from those on another, when all the collections are viewed as a whole there appears to be an almost continuous transition from one type to another. The following categories are therefore somewhat arbitrary and are not to be regarded as firm divisions.

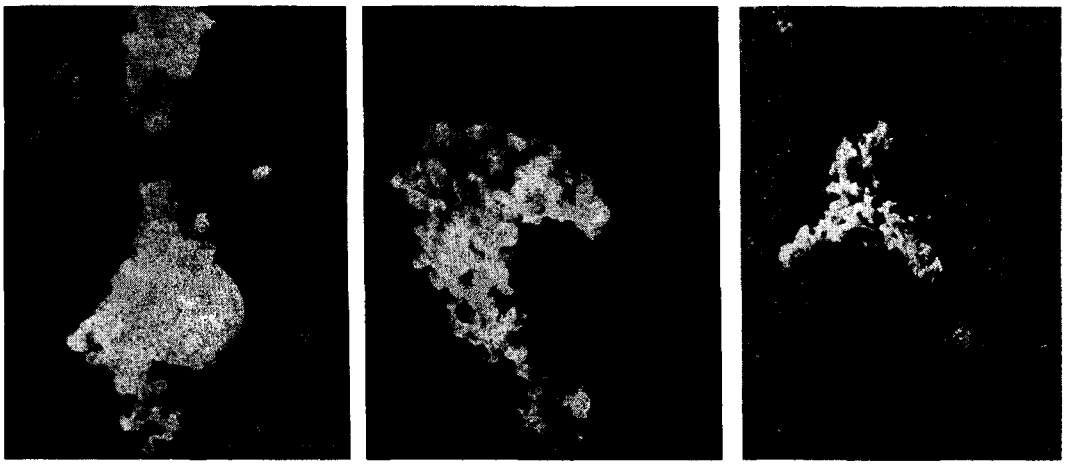

Fig. 4. See Fig. 3. Note liquid surrounding central particles in left and central photographs.

(1) Intricate chains of very small particles, shown in Figs. 3 and 4.

(2) More compact aggregates of very small particles (Fig. 5).

(3) Single crystals or small groups of relatively large crystals (Figs. 6 and 7) including nearly spherical particles.

(4) Irregular solids, usually fragile and showing crystalline faces or protrusions (Fig. 8).

These types are discussed in more detail below.

1. *Chains*

These show considerable variation in detail but an example which is typical of our collections has been clearly illustrated by Hemenway et al. (1961). The six photographs of Fig. 3 have been arranged to show the gradation from chains of small particles showing some well-marked crystalline (hexagonal) faces on the left, to more compact structures on the right which seem to have become cemented into a more solid mass. The gradations in the degree of darkness of the particles should be noted; it shows that they are partially transparent to electrons. This immediately rules out the possibility that they are metallic, or compounds of the heavier elements.

They are generally very stable under examination (except for liquids associated with some of them) even with intense electron beams.

The "Y" structure seen in two of these

Fig. 5. Compact aggregates of very small particles, surrounded by large stains.

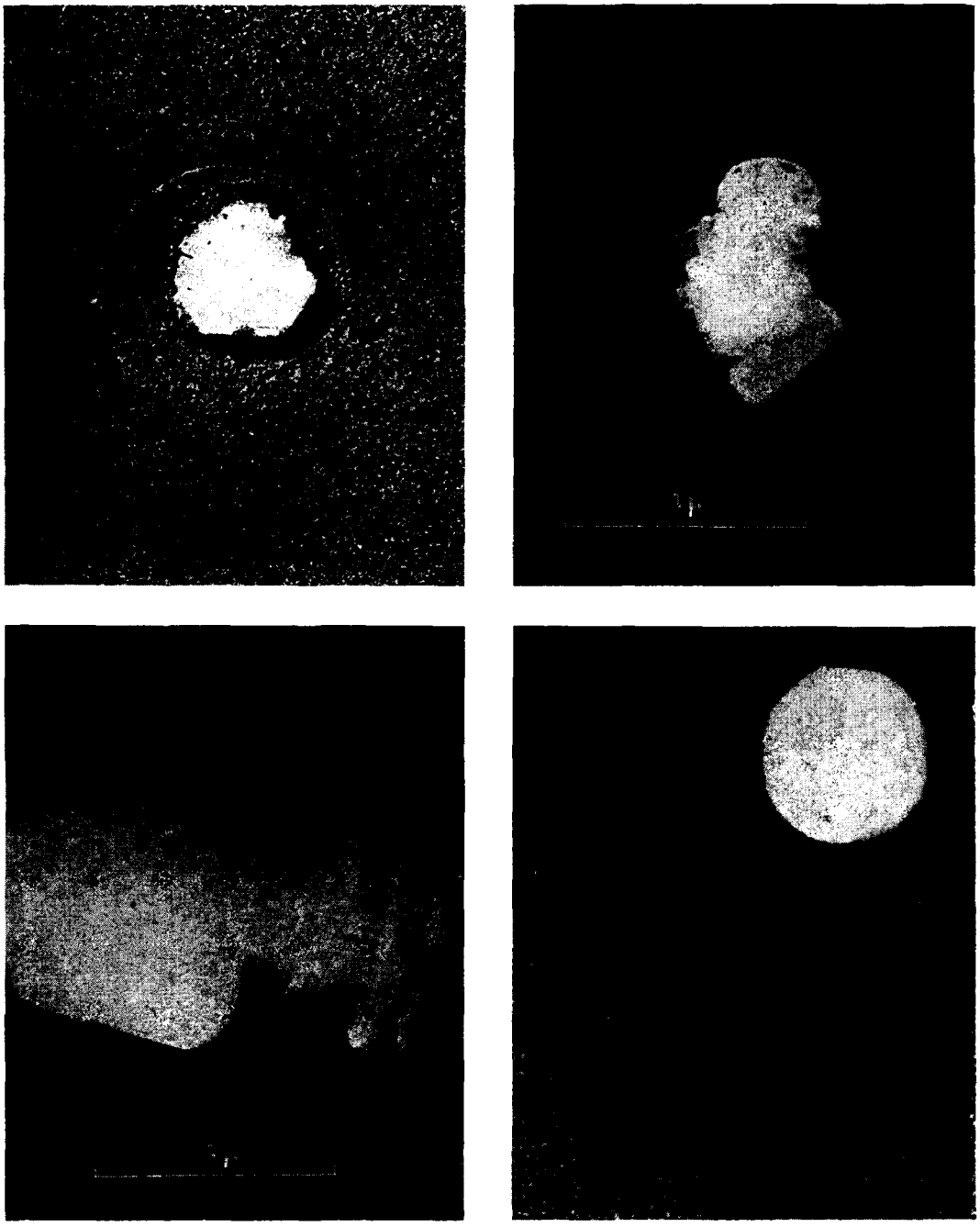

Fig. 6. Atmospheric particles with crystalline structure.

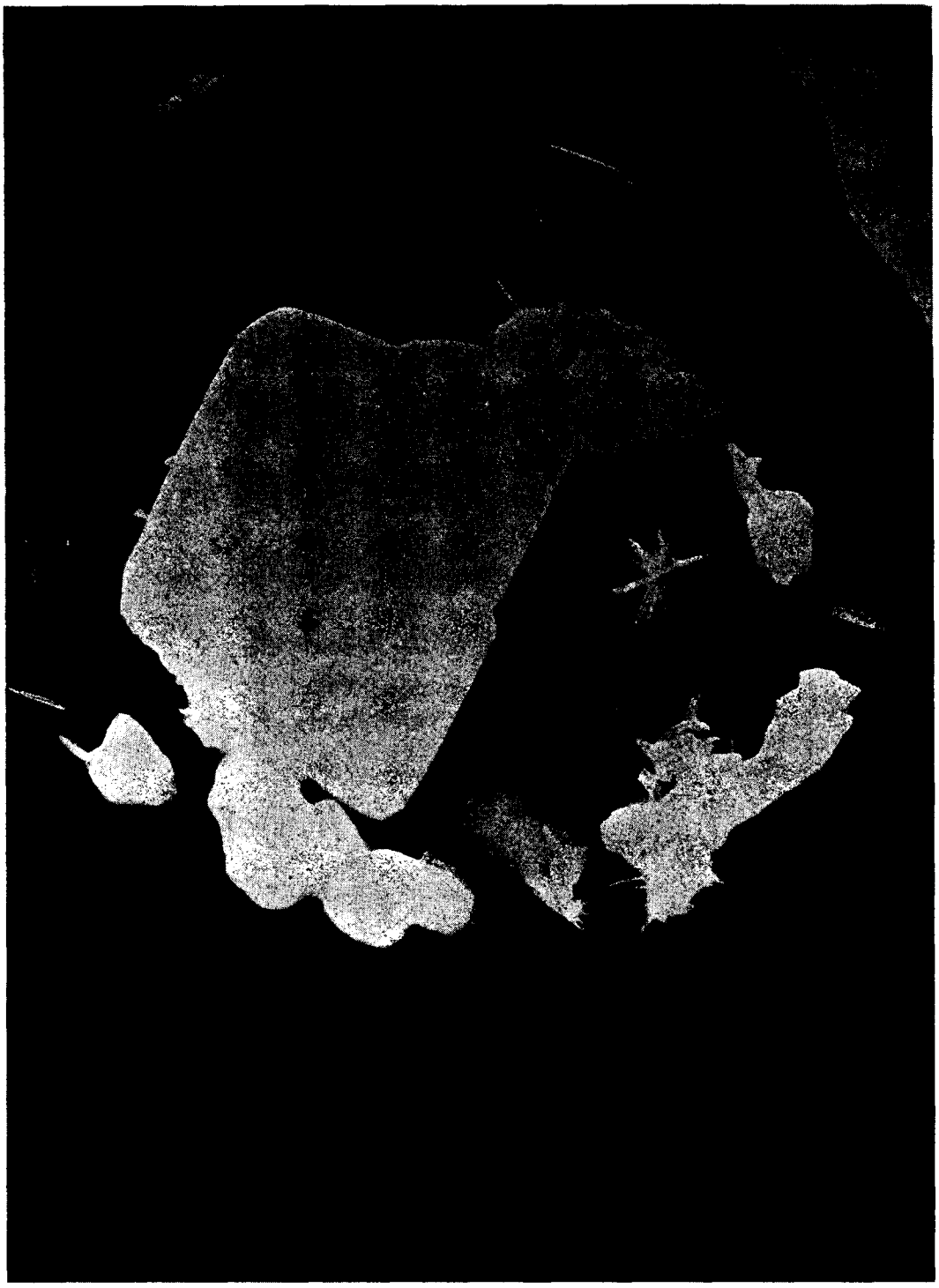

Fig. 7. The largest crystalline group of atmospheric particles.

Fig. 8. The remains of an impact of a large particle collected by a "whirling arm" impactor.

examples is quite common and is usually a tetrahedral arrangement. This gives the particles a very large total area compared with their area of contact with the surface, making them vulnerable to migration in the horizontally directed airstream on the impaction surface. The occurrence of such particles outside the impaction spot at first led us to believe that they were carbon chains, which superficially they resemble closely, collected during preparation of the screens for examination. However their failure to appear on unexposed screens, their association with volatile liquids which stain the surface or react with metals and the presence of crystalline facets which are not typical of carbon chains eventually demonstrated conclusively that they were collected at high altitude.

Sizes of the individual crystals of the group may vary from about 0.005 μm (50 Å) diam. to about 0.1 μm. The largest unbroken chain which we have obtained covered about 6 μm but some which broke on impact, if joined end to end would have exceeded 20 μm in length. When all particles of this type are recorded the most common length is about 0.3 μm but this probably bears no relation to their size before capture.

In Fig. 4, the appearance of some chains which have become wet by an associated liquid is shown in the first two photographs. Note that they have collapsed, losing their vertical structure; this appears to be common even with much smaller amounts of liquid, which must lubricate the joints.

There are apparently different types of associated liquids. As we stated earlier, use of a copper-coated nitrocellulose collecting surface leads to a marked reaction by chains which appear dry when collected on the usual carbon film. The photograph on the right of Fig. 4 shows a transition to the next particle type to be discussed. Note that the chain is much more compact than those of Fig. 3 and some associated material has caused the goldpalladium deposit near it to crack. This type of rather subtle reaction with the collecting surface is found with a minority of the chains but with almost all the compact aggregates. It suggests that the difference is mainly due to the associated liquids.

A few chains have been collected which show evidence of a small amount of sulphuric acid on them, while others collected at lower altitudes have been found immersed in it without visible reaction other than the loss of their rigidity. On the other hand there appear to be far fewer below 25 km than above, so that many must dissolve when subjected to sulphuric acid immersion. This suggests that some chains may be quite different chemically from others. Because we have also found chains consisting of both spherical particles and crystalline forms, any one chain is not necessarily homogeneous.

2. *Compact aggregates of small particles*

Figure 5 shows the next stage from the transition particle of Fig. 4. Each of these three particles, although less than 1 μm in diameter is surrounded by an area of roughly concentric stains exceeding 10 μm in diameter, in which the shadowing deposit is of variable density and texture. Although all the material causing these variations has usually disappeared, occasionally enough remains to give the appearance of a very thin liquid layer which has crystallized into hexagonal shapes. In the left photograph this is visible on the original plate, and some of the layer is clearly seen in the immediate vicinity of the particle.

Aggregates as small as 0.1 μm are still accompanied by enough of this volatile material to form stains several microns in diameter.

The size of the individual components of the aggregates are generally more uniform and smaller than for the chains, (0.005–0.015 μm) (50–150 Å) being typical. The centre photograph shows some of them scattered around the parent body.

3. *Single crystals or groups of crystals*

Some examples of these are shown in Fig. 6 and may not really be distinct from the preceding types except because of their larger size and isolation. Many of them look suspiciously like rhombohedral or pseudohexagonal twinned forms of ammonium sulphate. Note that the lower right-hand photograph, at first sight a sphere, has on closer examination crystal facets. This appears to be common to many of the spheres we have observed. True spheres exist, both with and without associated liquid, but are quite rare.

Fig. 7 shows the largest crystalline group yet obtained. Obviously the associated liquid has evaporated and the solids have crystallized out.

4. *Irregular large solids*

Most of these, like the large crystalline groups, have come from the "whirling arm" impactor, for the sedimentation chamber used in the other impactor discriminates against large particles. The group in Fig. 8 is typical and from the emptiness of the surrounding screen was clearly the remains of one large particle. It impacted on the surface at 16 m sec^{-1} but in spite of its relatively large size only dented the 500 Å thick nitrocellulose film instead of penetrating it and then shattered into many pieces. It should be noted that the largest portion has had some liquid component associated with it which has left a stain ring around it. The individual pieces are quite typical of the irregular particles collected, which nearly always show crystalline facets or protrusions, and some straight edges. Even large pieces are often partially transparent to the electrons.

Larger particles than this usually break the nitrocellulose film and some have left rather spectacular collections of chains, crystal groups and stains around the edge of the hole. Evidently all the types we have described may be associated in one body and their separation may have occurred on entering the atmosphere.

Discussion and Conclusions

The fragile partially electron-transparent crystalline particle groups and their associated liquids which we have found are completely different from the micrometeorites conventionally sought in polar ice and deep-sea deposits. They are however entirely in character with the current notion of cometary debris. If this is their origin it is clear that in the micron and submicron particle range the input of this material to the earth must far outweigh the contribution from the nickel-iron and silicate types for electron dense particles are in a very small minority in this size range.

In one of the most comprehensive and relevant papers published recently on the influx to the earth of cometary material, Silverberg (1970) has made several important points.

One is that the occasional high flux rates of microparticles measured by acoustic detectors on satellites (which have been largely discredited by the failure of penetration experiments to find comparable rates) could be explained if the particles concerned had a large area-to-mass ratio (i.e. low effective density). He further went on to show that the times of occurrence of all such events could be adequately explained in terms of the expected positions of dust near the orbits of certain comets, provided again that the particles had this low effective density. A glance at Fig. 3 shows that this type of particle would have just the characteristics required, while the partially liquid nature of many others may have rendered them more compact than they would have been in the low temperatures of space.

Another point made by Silverberg is that submicron particles (which include many of our single crystals) are so close to the radiation pressure limit which would exclude them from the earth's environment, that it is unlikely that they have entered the atmosphere in their present form. It is easy to imagine that if they were originally attached to any of the other particle types we have illustrated, they would become detached on entering the atmosphere. The true cometary particles may therefore be assemblages of the particles which we have illustrated.

We have not yet obtained sufficient samples at about the same time in successive years to be sure that distinctive particle types are an annual event, but our most interesting results have been obtained in March 1968, 1969 and 1970. However, the particles on each occasion were quite different so that if they belong to the February cometary streams associated with comets Brorsen-Metcalf or Encke, as discussed by Silverberg, then there has been considerable differentiation of particle types around the orbit.

Silverberg states that the cohesive force between small grains is so large that apparently fragile structures could easily survive the forces present in a comet. Since the particles of Fig. 3 collided with the collecting surface at not less than 100 m sec^{-1}, with only occasional fractures, his argument is certainly reasonable.

It is possible in principle to calculate the flux of incoming particles in a given size range from the concentrations which we obtained, assuming a stagnant atmosphere and that the

fall velocity is known. Unfortunately the fall velocities are so low the atmosphere may not be "stagnant", the fall velocity is very difficult to calculate for the odd-shaped particles, many particles may break up to form smaller ones, we do not know the shapes or sizes in the atmosphere of particles which are liquid when examined at room temperatures while others may be blown off the impaction surface or evaporate before examination. Any estimate will therefore be very crude. Taking the values given by Bigg, Ono & Thompson (1970), at 32 km we find typically about 4.10^4 m^{-3} of particles equivalent in median size to spheres of 0.3 μm diameter. The fall velocity is then of the order of 3.10^{-4} m sec^{-1} (for a density of 1) and the mass 10^{-14} gm. The flux is then about 10 m^{-2} sec^{-1} but, of course, subject to the large possible errors mentioned.

This value is intermediate between the highest and lowest estimates from rockets and satellites. Because we have found so much material of a volatile nature it is evidently very important to have a cold surface for impaction both during and after collection. Our sampling is carried out at temperatures colder than −30°C but the surface may subsequently warm to 20° or 30°C while awaiting recovery. Storage is normally at 20°C until the collection is shadowed. Surfaces exposed from rockets may be subjected to considerable aerodynamic heating, particularly when opened below 80 km and the different flight parameters in different experiments may account for some of the very large discrepancies which have been recorded in particle fluxes.

To sum up, even though in our experiments the particles are subject to alteration by fragmentation and evaporation in the atmosphere and in our collectors and possibly to chemical changes in the atmosphere, it seems that we may have here the best available method of examining cometary constituents. Clearly we have made only a small beginning with this work; there is so much that could be done to improve our collection methods (for example by keeping the sample cold), to identify the chemicals involved and to survey the annual changes in particle appearance, concentration and composition. We hope that these results will stimulate other groups to participate in what will otherwise be a very long task and will also be useful to those searching for extraterrestrial particles by any other means.

Acknowledgements

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ФОТОГРАФИИ ВНЕЗЕМНЫХ ЧАСТИЦ, ПОЛУЧЕННЫЕ С ПОМОЩЬЮ ЭЛЕКТРОННОГО МИКРОСКОПА

С помощью соответствующих методов сбора можно отличить частицы, существующие в стратосфере, от загрязнений и выделить частицы внеземного происхождения. Представлен каталог портретов частиц под электронным микроскопом, который показывает наиболее часто встречающиеся типы таких частиц по материалам около 40 коллекций, собранных на аэростатах в течение трех лет. Мы делаем заключение, что малые внеземные частицы наиболее часто встречаются в виде хрупких скоплений низкой плотности ещё более мелких частичек, часто составленных из кристаллического материала и связанных с жидкими или твёрдыми летучими компонентами. Предполагается, что последние имеют кометное происхождение.