

Simulation of ring patterns observed with noctilucent cloud particles

By R. A. SKRIVANEK and R. K. SOBERMAN, *Air Force Cambridge Laboratories, Bedford, Mass.*

(Manuscript received December 15, 1963)

ABSTRACT

Laboratory simulation of the ring or halo patterns which were found in the electron microscopic examination of noctilucent cloud particles has been attempted. This was done by impacting ice coated nickel particles on collecting surfaces similar to those used in the 1962 rocket sampling of the clouds. Ring patterns similar to those observed on the rocket sampling surfaces have been produced. From the condition necessary to produce these patterns, a formation mechanism is inferred.

Introduction

Electron microscopic examination of the noctilucent cloud particles collected during 1962 in Sweden (Ref. B) revealed an unusual ring, or halo structure surrounding many of the particles (see Fig. 5, Ref. B). Speculation arose as to the nature of these rings and possible mechanism responsible for their formation. As the investigation of the collected particles proceeded evidence was found indicating that the particles were probably surrounded by a coating of water or ice at the time of collection. (Ref. D). In an attempt to explain the ring structure, a series of laboratory experiments were performed to simulate these rings.

Experimental procedure

For applying the particles to the surfaces in these simulation experiments a spray mounter was used. A capillary tube containing the desired particles was inserted into the sprayer, and a stream of high purity liquid, or gaseous Freon was used to propel the particles toward the target (see Fig. 1). The particle collection surface consisted of 200–300 Å thick nitrocellulose film, with a 50 Å thick evaporated coating of aluminum. The particles were sprayed on the nitrocellulose from a distance of 20 to 30 cm. Where it was necessary to control the humidity the particle spray mounting was carried out in a chamber which was designed for this process.

When a slurry of particles in water was sprayed using gaseous freon as the propellant, the water impacted the nitrocellulose as a liquid. By inverting the spray mounter, liquid freon was obtained for the propellant. Under those conditions the water froze and impacted as ice. After the surfaces had been sprayed, they were placed in a dust free hood for about 1 hour. This allowed sufficient time for the moisture from the spray to evaporate. The surfaces were then put into a vacuum evaporator, shadowed with aluminum at an angle of approximately 15° to the surface and subsequently examined in an electron microscope. The aluminum shadowing was done to aid in the microscope observations (as described in Ref. B).

Results and conclusion

Optimum results were obtained by using liquid freon to propel sub-micron nickel particles which had been suspended in distilled water. The ring structures obtained in this fashion showed certain similarities to the noctilucent cloud particle rings. Fig. 2a is an electron micrograph of a noctilucent cloud particle. Within the ring, minute particles can often be seen as well as what appears to be a staining or distortion of the nitrocellulose substrate. These same characteristics are evident in the simulated ring (Fig. 2b).

JUNGE and MANSON (1961) found particles surrounded by ring structures in several high

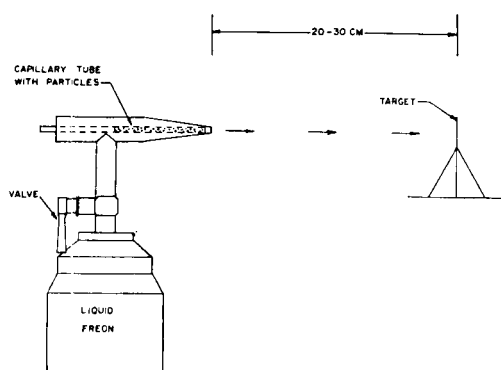


FIG. 1. Freon gas spray mounter used for the simulation experiment.

altitude U-2 collections. They attributed the rings to water vapor which had contaminated the collection surfaces. Many of their particles, containing ammonium sulfate, were shown to be hygroscopic. When the relative humidity was raised to a point between 72 and 80 % these particles would absorb water and dissolve. If the relative humidity was subsequently lowered to about 40 % the particles would recrystallize. Further, a change in the particle structure could be observed in the electron microscope as the beam illuminated the particles. Water evaporated from the particles and left a central nucleus surrounded by a ring structure. Thus these authors attributed the ring structure to water vapor contamination of their collection. When sampling London fog with a Casella thermal precipitator, WALLER, BROOKS and CARTWRIGHT (1963) also reported finding ring structures on formvar coated glass slides. They

conducted laboratory simulation experiments and arrived at the conclusion that the rings were formed by aerosols of salts and sulfuric acid.

The applicability of the conclusions of Junge and Manson to the present collection experiments were tested. Noctilucent cloud particles on collection surfaces from the rocket flight were subjected to 100 % relative humidity at sea level pressure and a temperature of approximately 0°C for one hour and then placed immediately in an electron microscope for examination. Neither the particle nor the ring had changed appearance and no amount of heating by the electron beam could produce a change. Such a change would be expected if the rings were due to hygroscopic particles.

Further, it is to be noted that no ring structures were found on the control surfaces that shared the same environment with the collection surfaces on the rocket (Ref. B) which would have been the case if these structures were the result of contamination by water vapor. Thus, one concludes that the ring structures associated with the noctilucent cloud particles cannot be explained in the same fashion as those associated with the ammonium sulfate particles reported by Junge and Manson, or by the sulfuric acid particles of Waller *et al.*

The present simulation studies were conducted with nickel particles which are non-hygroscopic. When the humidity tests reported above were tried with these particles, as with the noctilucent cloud particles, no changes were observed. It was observed, however, that humidity does influence the formation of the

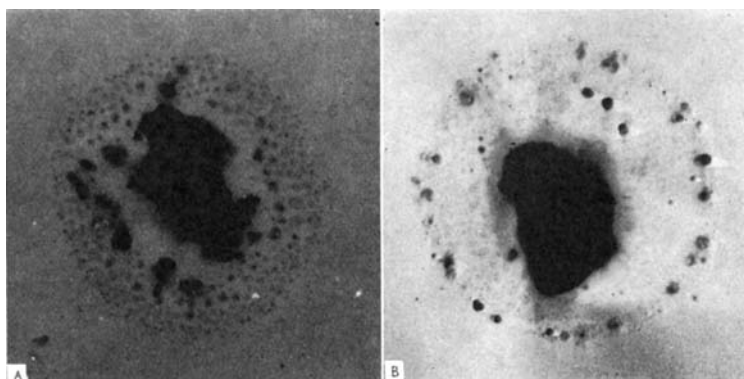


FIG. 2. (a) Electron micrograph of noctilucent cloud particle showing ring structure. (b) Electron micrograph of laboratory simulated ring structure. Scale = 1 micron.

TABLE 1. *Effect of relative humidity on ring formation.*

Relative Humidity During Spray Mounting %	Relative Humidity During Drying %	Percentage of Particles with rings to the total %
50	50	0
70	36	0.1
82	32	0.1
90	44	0.1
36	70	0.1
38	77	0.1
70	70	0.1
77	77	Between 1 and 10
82	82	Between 1 and 10
90	90	Between 1 and 10
40	80	Between 0.1 and 1
32	82	Between 0.1 and 1
44	90	Between 1 and 10

ring structure in these simulation experiments. The humidity was varied during the spray mounting process and during the subsequent drying period before the low angle evaporated aluminum coating was applied. The results are shown in Table 1. From this table it can be seen that: 1) if the relative humidity was below 70 %, less than 1 % of the particles had any surrounding ring structure. 2) at 50 % no ring structures were seen. 3) The high humidity is most effective on the process of ring formation after the initial impactation and during the time that the moisture on the surface is evaporating.

To explain the formation of the ring patterns the following hypothesis is offered: If an ice coated particle impacts on a relatively warm nitrocellulose film in an environment where the relative humidity is high, the ice melts and the water remains around the particle for a relatively long time. This enables the water to stain and deform the film before evaporating. If the relative humidity is low, the ice sublimates

or evaporates rapidly and has little effect on the film. Further, the particles that are sometimes seen in the ring structure would be drawn to the central nucleus if the evaporation rate was high, whereas in a slower process these small particles could settle to the film and be deposited in the form of a broader ring surrounding the central larger particle. Presumably the coating must be in the form of ice to maintain a cold temperature in the vicinity of the particle. The small particles may be fragments of the central nucleus or they could be the result of agglomeration during the time the ice coating was being formed.

In conclusion, it is believed that these experiments have been able to simulate some of the features of the ring patterns that were found on the noctilucent cloud particle collecting surfaces. That the appearance is not exactly the same is attributed to the difference in the environmental conditions (i.e. impact velocity, temperature and pressure). Further experiments are planned where these environmental factors will be stimulated. However, the evidence of these simulation experiments when considered with the results of the calcium collection surfaces that were carried on the rocket flights, (Ref. D) leads to the conclusion that the ring patterns which were found associated with many of the noctilucent cloud particles were caused by ice coatings which surrounded those particles in situ. This conclusion has been given additional support by recent temperature measurements in the clouds. The measurements show an exceedingly low temperature in the clouds and are not inconsistent with a super-saturation condition that would lead to condensation in the form of ice coatings on particles.

Acknowledgements

We wish to thank C. L. Hemenway, G. Witt and N. Wilhelm for many helpful discussions and suggestions and Miss L. Cohen for her technical assistance.

REFERENCES

- JUNGE, C. and MANSON, J. E., 1961, Stratospheric Aerosol Studies. *J. of Geophysical Research*, Vol. 66, p. 2163.
- WALLER, R. E., BROOKS, A. G. F. and CARTWRIGHT, J., 1963, An electron microscope study of particles in Town Air. *Int. J. Air Wat. Pol.* 7.
- A. Techniques for Rocket Sampling of Noctilucent Cloud Particles, by R. K. SOBERMAN, S. A. CHREST, J. J. MANNING, L. REY, R. A. SKRIVANEK and N. WILHELM. *Tellus*, 16, 1.
- B. Electron-microscopic Studies of Noctilucent Cloud Particles, by C. L. HEMENWAY, E. F. FULLAM, R. A. SKRIVANEK, R. K. SOBERMAN and G. WITT. *Tellus*, 16, 1.
- C. Composition Analysis of Noctilucent Cloud Particles, by G. WITT, C. L. HEMENWAY, N. LANGE, S. MODIN and R. K. SOBERMAN. *Tellus*, 16, 1.
- D. Calcium Film Indicator of Moisture Associated with Noctilucent Cloud Particles, by I. LINSCOTT, C. L. HEMENWAY and G. WITT. *Tellus*, 16, 1.