LIGHT SCATTERING BY NONSPHERICAL ICE GRAINS: AN APPLICATION TO NOCTILUCENT CLOUD PARTICLES

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Abstract. The light scattered by noctilucent cloud particles is nearly fully polarized at scattering angles in the vicinity of 90°. This was one of the reasons to conclude that the upper limit of their sizes is not larger than about 0.12 μm. Nevertheless, this estimate was made on the basis of the Mie scattering theory for spherical particles, whereas many investigators noted usefulness of highly aspherical shapes of noctilucent cloud particles. In this paper, we used rigorous light scattering theory for randomly oriented nonspherical particles to calculate the degree of linear polarization of the scattered light for ice grains of different shape. By comparing these calculations with rocket polarization measurements of noctilucent clouds, we show that, as for spherical particles, the upper limit of particle equal-volume radii for slightly flattened and elongated grains is of about 0.12 μm, while for highly aspherical plate-like and needle-like particles this upper limit is substantially larger and is of about 0.18–0.20 μm. We also report calculations of the volumetric scattering cross-section for particles of different shape and show that randomly oriented spheroids have (slightly) smaller scattering cross section per unit particle mass than equal-volume spherical grains. Nevertheless, if in noctilucent clouds plate-like and needle-like grains grow to much larger sizes than spherical particles, their scattering efficiency may be much greater.

1. Introduction

Determination of sizes of noctilucent cloud particles by means of interpretation of photometric and polarimetric observations has been the subject of many publications (see, e.g., Table 1 of Thomas and McKay, 1985), but no definite consensus has been achieved yet (e.g., Gadsden and Schröder, 1989). Though the particles of noctilucent clouds are by no means spherical in shape, most of estimates of their size are based on the Mie scattering theory. On the other hand, some authors have pointed out the usefulness of highly aspherical particle shapes in producing larger particles due to (much) smaller settling velocities and thus in maximizing the scattering efficiency while minimizing the required water content (e.g., Reid, 1975; Gadsden, 1977; Garcia, 1989). It follows form computations of Wiscombe and Mugnai (1986) that small, nearly spherically shaped Chebyshev particles have scattering properties very much like those of equal-volume spheres. Nevertheless, this may be not true for highly aspherical needle-like and plate-like scatterers.

It is well known that, generally, polarization measurements contain more reliable information about particle size and shape than photometric ones. In an important paper, Bohren (1983) used Rayleigh scattering theory for very small ellipsoids to analyse polarimetric observations of noctilucent clouds. On the one hand, Bohren argued that, though particles with radii of about 0.1 μm at visible wavelengths are beyond the limit where Rayleigh theory is strictly valid, there is

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no sharp cutoff beyond which Rayleigh theory gives completely invalid results, and, thus, this theory may be extended slightly beyond its purview. On the other hand, he pointed out that Rayleigh theory predicts no substantial differences between light scattering properties of spherical and randomly oriented nonspherical particles. Therefore, Bohren suggested that the Mie scattering theory for spheres may be used to infer the size of noctilucent cloud particles provided that this size is not too large as compared with that allowed by Rayleigh theory. As a result, Bohren concluded that, whatever the shape of noctilucent cloud particles is, the upper limit of their sizes cannot be much larger than about 0.1 μm.

The aim of the present paper is twofold. First, we use rigorous light scattering theory for randomly nonspherical particles of arbitrary size to reexamine Bohren's conclusions. Specifically, we rigorously compute polarization properties of spheroidal ice grains and, by comparing these calculations with polarimetric observations of noctilucent clouds at scattering angles near 90°, determine how the upper limit of particle sizes depends on particle asphericity. Second, we calculate volumetric scattering cross sections for particles of different shape and discuss whether highly aspherical particle shapes are really advantageous in producing the maximum scattering efficiency while minimizing the required water content, as is usually suggested.

2. Calculations: Polarization

Following Bohren (1983), we assumed that noctilucent cloud particles are randomly oriented and, thus, their scattering properties may be characterized by 4×4 scattering matrix $\mathbf{F}$ of the form

$$\mathbf{F} = \begin{bmatrix} F_{11} & F_{12} & 0 & 0 \\ F_{12} & F_{22} & 0 & 0 \\ 0 & 0 & F_{33} & F_{34} \\ 0 & 0 & -F_{34} & F_{44} \end{bmatrix}, \tag{1}$$

where F_{ij} are some real-valued functions of the scattering angle. For unpolarized incident light, the degree of linear polarization of the scattered light is defined as

$$P = -F_{12}/F_{11}\,. \tag{2}$$

We modeled nonspherical particle shapes by oblate and prolate spheroids. The

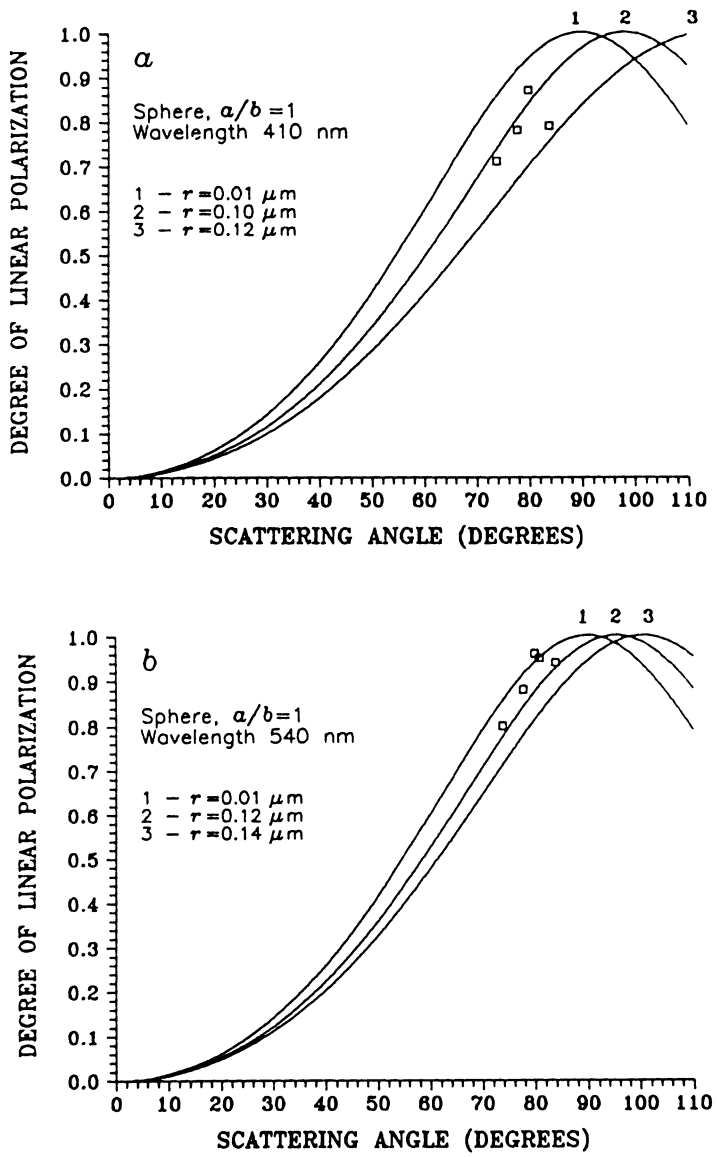

Fig. 1. Computed degree of linear polarization of the scattered light versus scattering angle for ice spheres. Boxes show observational data of Tozer and Beeson (1974).

shape of a spheroid in its natural spherical coordinate system is given by

$$r(\theta, \varphi) = a\left(\sin^2\theta + \frac{a^2}{b^2}\cos^2\theta\right)^{-1/2}, \tag{3}$$

where b is the rotational semi-axis, and a is the horizontal semi-axis. Another pair of parameters, that may be used to specify the shape of the spheroid, is (r_{ev}, d),

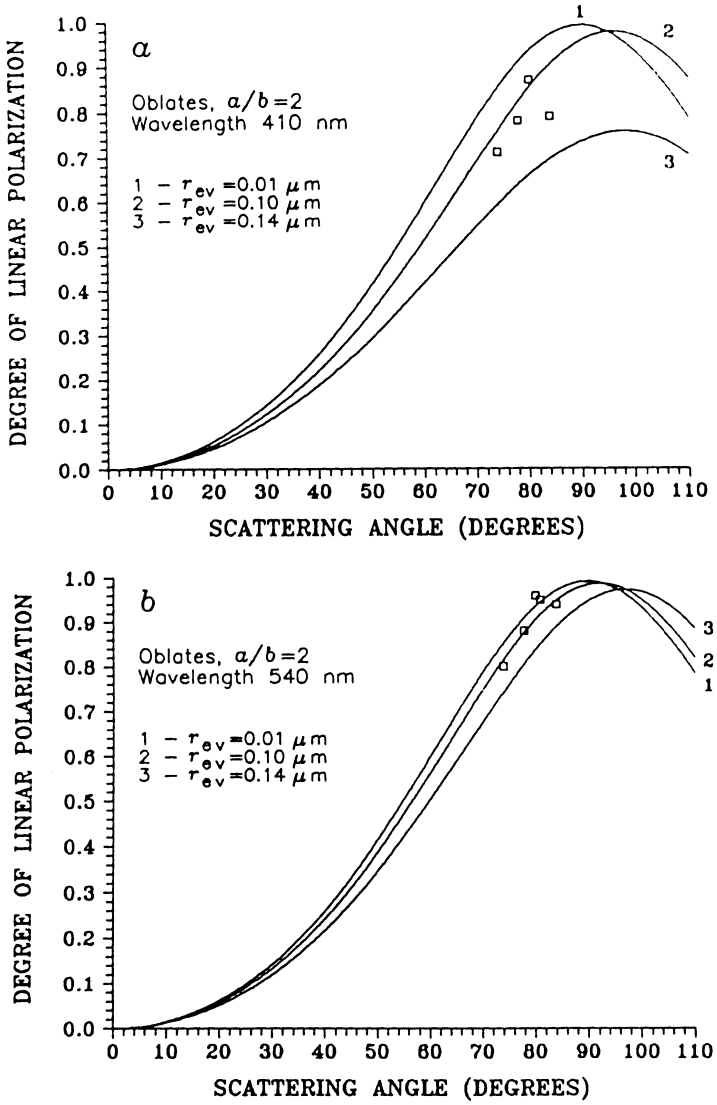

Fig. 2. As in Figure 1, for randomly oriented oblate spheroids with $d = 2$.

where $d = a/b$ is the ratio of the semi-axes, and r_{ev} is the radius of the equal-volume sphere given by

$$r_{ev} = a\, d^{-1/3}\,. \tag{4}$$

Note that $d > 1$ for oblate spheroids and $d < 1$ for prolate spheroids.

To calculate the degree of linear polarization of the scattered light for randomly oriented spheroidal grains, we used the rigorous light-scattering theory developed by Mishchenko (1991). Since polarization at scattering angles near 90° is most sensitive to particle size and shape, we analysed rocket measurements of Tozer

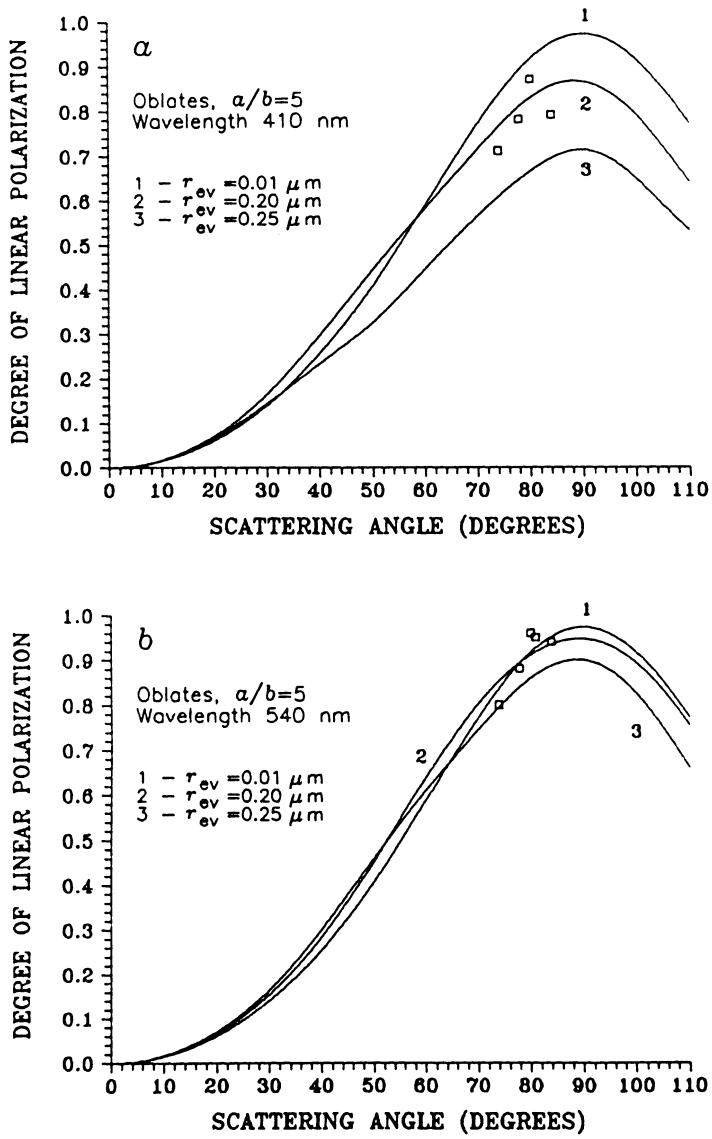

Fig. 3. As in Figure 2, for $d = 5$.

and Beeson (1974), which refer to scattering angles 74 to 84 degrees and wavelengths $\lambda = 410$ and 540 nm. As usually, we assumed that noctilucent cloud particles are composed of ice and used refractive indices 1.32 ($\lambda = 410$ nm) and 1.31 ($\lambda = 540$ nm) (Warren, 1984).

Results of our computer calculations for spherical and randomly oriented spheroidal ice grains are shown in Figures 1–5. One sees that for spherical and slightly flattened and elongated spheroidal grains the upper limit of equal-volume radii is about 0.12 μm, in agreement with earlier estimates of Witt (1960) and Tozer

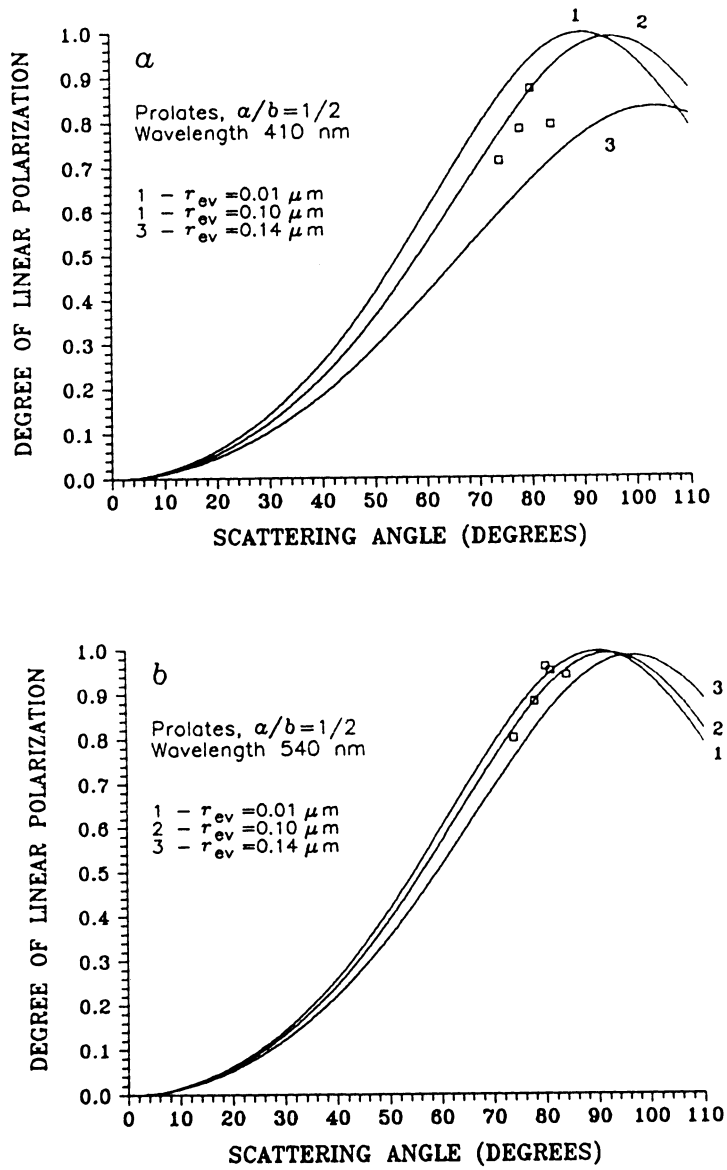

Fig. 4. As in Figure 1, for randomly oriented prolate spheroids with $d = 1/2$.

and Beeson (1974). Nevertheless, for highly aspherical plate-like and needle-like spheroids with $d = 5$ and 1/5, respectively, the upper limit of equal-volume radii is substantially larger and is of about 0.18–0.20 μm. As was noted by Reid (1975), highly aspheric grains have considerably slower settling velocities and, therefore, are able to grow to larger sizes than spherical grains. Our calculations show that these greater sizes are admitted not only by cloud microphysics, but also by cloud polarimetry.

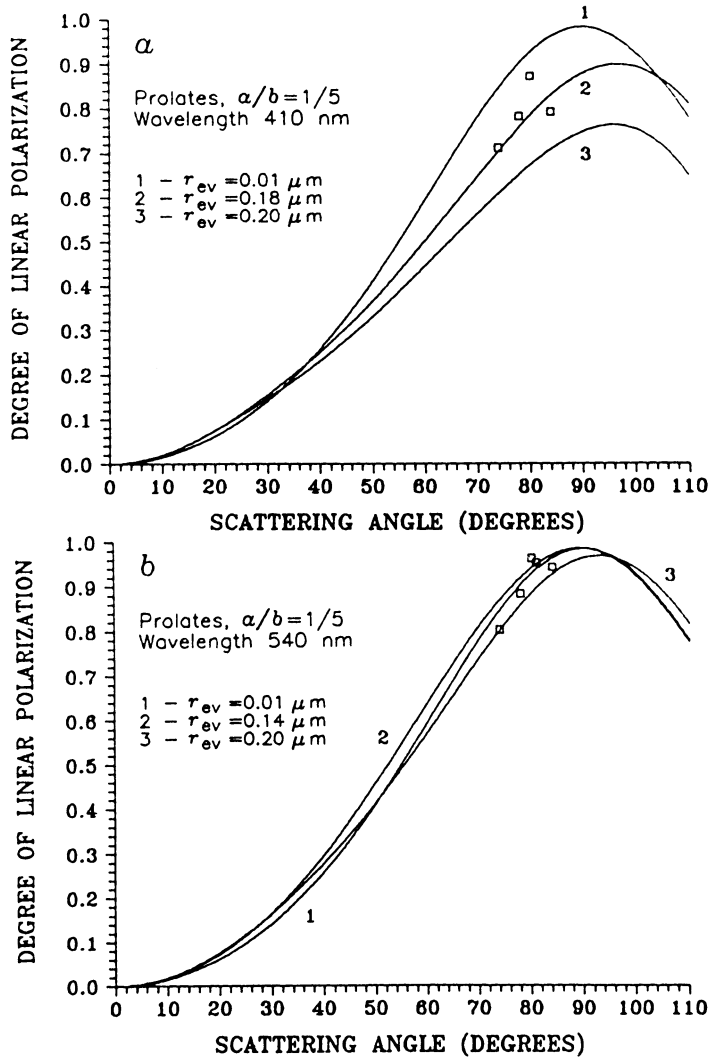

Fig. 5. As in Figure 4, for $d = 1/5$.

3. Calculations: Scattering Cross-Section per Unit Particle Volume

To give the observed brightness of noctilucent clouds, some amount of ambient water vapor is necessary which critically depends on possible particle size and shape. First, as follows from light scattering theory, the larger the particles, the smaller mass of these particles is required for a given brightness. Second, if the particles are spherical, then their settling velocities are large and their residence time in the region of supersaturation is small. Therefore, too great ambient amount of water vapor may be required to achieve sufficiently fast growth rates (Garcia, 1989). On the other hand, highly aspheric plate-like and needle-like particles have much slower settling velocities (Reid, 1975) and, therefore, are able to grow to

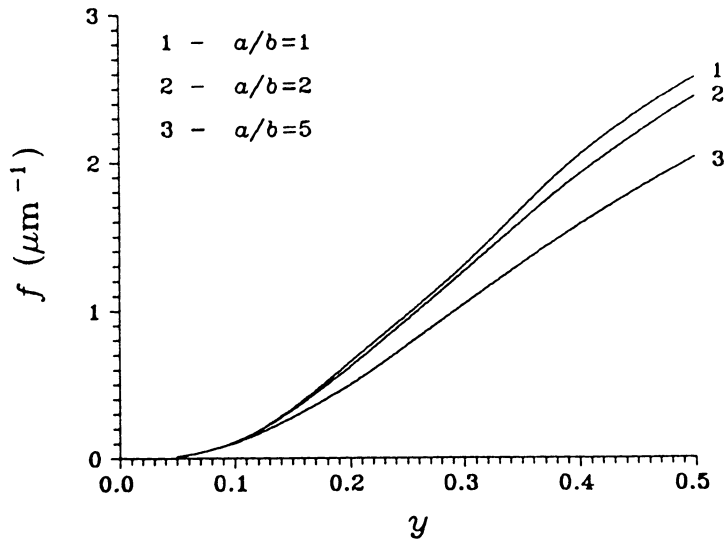

Fig. 6. Scattering cross-section per unit particle volume $f = C_{sca}/(\frac{4}{3}\pi r_{ev}^3)$ versus a nondimensional parameter $y = r_{ev}/\lambda$ for ice spheres and randomly oriented oblate spheroids with $d = 2$ and 5. Computational results for prolate spheroids with $d = 1/2$ and 1/5 nearly coincide with those for oblate spheroids with $d = 2$ and 5, respectively. The refractive index is 1.31.

sufficiently large sizes even in the presence of small amounts of water vapor (Garcia, 1989). Thus, highly aspheric particle shapes seem to be advantageous in maximizing the scattering efficiency while minimizing the required water content.

The scattering cross-section per unit particle volume f is given by

$$f = C_{sca}/(\tfrac{4}{3}\pi r_{ev}^3)\ , \tag{5}$$

where C_{sca} is the scattering cross-section. In Figure 6, computed values of f are plotted versus a nondimensional parameter $y = r_{ev}/\lambda$ for particles of different shape. One sees that randomly oriented spheroids have (slightly) smaller scattering efficiency per unit mass than equal-volume spheres. Nevertheless, Figure 6 evidently demonstrates that if, due to particular microphysical conditions, highly aspherical particles grow to much greater sizes than spherical grains, their scattering efficiency may be much larger.

4. Summary and Conclusions

High degree of linear polarization of the scattered light observed for noctilucent clouds is usually interpreted as an evidence for very small sizes of cloud particles. In this paper, we have used rigorous light scattering theory for randomly oriented nonspherical particles of arbitrary size to analyse rocket polarimetric observations of Tozer and Beeson (1974), which refer to the most informative range of scattering angles near 90°. We have found that the upper limit of particle sizes strongly

depends on the particle shape and for plate-like and needle-like particles is 1.5–2 times that for spherical grains. This result may be of importance in microphysical modeling of noctilucent cloud particles. Also, we have calculated volumetric scattering cross sections for particles of different shape and concluded that if in the range of noctilucent cloud formation plate-like and needle-like particles grow to much greater sizes than spherical particles, their scattering efficiency may be much larger.

References

Bohren, C.: 1983, *Tellus B* **35**, 65.
Gadsden, M.: 1977, *Ann. Geophys.* **33**, 357.
Gadsden, M. and Schröder, W.: 1989, *Noctilucent Clouds*, Springer-Verlag, Berlin.
Garcia, R. R.: 1989, *J. Geophys. Res. D* **94**, 14605.
Mishchenko, M. I.: 1991, *J. Optical Soc. Amer. A* **8**, 871.
Reid, G. C.: 1975, *J. Atmos. Sci.* **32**, 523.
Thomas, G. E. and McKay, C. P.: 1985, *Planet. Space Sci.* **33**, 1209.
Tozer, W. F. and Beeson, D. E.: 1974, *J. Geophys. Res.* **79**, 5607.
Warren, S. G.: 1984, *Appl. Optics* **23**, 1206.
Wiscombe, W. J. and Mugnai, A.: 1986, *Single Scattering from Nonspherical Chebyshev Particles: A Compendium of Calculations*, NASA Ref. Publ. 1157.
Witt, G.: 1960, *J. Geophys. Res.* **65**, 925.