Optical characteristics of mesospheric aerosol distributions in relation to noctilucent clouds

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ABSTRACT

It has not hitherto been possible to establish whether the appearance of noctilucent clouds is due to an increased concentration or locally increased size of solid particles at the mesopause, or to a combination of these mechanisms. In this contribution the possibility of a relationship between the optical characteristics of noctilucent clouds and ice sublimation is discussed on the basis of present-day knowledge of the structure and photochemistry of the mesosphere.

The first part is devoted to a discussion of the character of the sublimation process at these altitudes. It is found that such a process is possible in conditions prevailing at the high-latitude summer mesopause. The time scale of sublimation and evaporation is slow compared with horizontal translation velocities observed in this region. Below the mesopause evaporation becomes rapid compared with the settling speed of sub-micron particles.

In the second part the scattering properties of monodisperse and polydisperse particle distributions, and the possible changes following a sublimation process are reviewed on the basis of numerical computations of scattering functions of coated spherical particles. It is found that the optical consequences of sublimation depend critically upon the presence and relative concentration of particles obeying the Rayleigh scattering law in the visible spectrum. The characteristic time scale of brightness changes by sublimation is at least 10 to 100 minutes.

Different optical methods for further investigation of the nature of mesospheric aerosols are summarized and a method is proposed for the detection of scattering layers in the mesosphere by means of ultraviolet intensity and polarization measurements from rockets and satellites.

Introduction

A display of noctilucent clouds is from the point of view of optics a temporary enhancement of the atmospheric scattering coefficient confined to a distinct and relatively narrow altitude range in the vicinity of the mesopause. From measurements of the degree of polarization of the light scattered by the clouds (Witt, 1960; Willmann, 1962) and by analysis of cloud spectra (Deirmendjian & Vestine, 1958), it has been established that the cloud layer is formed by solid particles following some form of size distribution law, and further that the size of particles in an equivalent uniform-size aerosol would range from 0.1 to 0.4 microns depending on the measuring technique employed. During 1962 a co-operative Swedish-American attempt was made for direct collection of noctilucent cloud particles by means of sounding rockets (Hemenway, Soberman & Witt, 1964). The experiment was undertaken from the Kronogård rocket range in Northern Sweden (66° N). Results were obtained from two flights, one in the presence of noctilucent clouds and one when these were not observed. The electron-microscopical analysis of the collecting surfaces indicated a significantly increased concentration of solid particles in the presence of noctilucent clouds. The particles collected in the layer between 78 to 94 km were found to follow a cumulative size distribution law of the form $N \sim r^{-p}$, $(3 \leqslant p \leqslant 4)$ for radii greater than about 0.025 microns. Figure 1 shows electron micrographs of sub-micron particles collected during the cloud flight. The feature surrounding the particles a halo-like structure which was only observed in the noctilucent cloud samples, has been interpreted as possibly being due to

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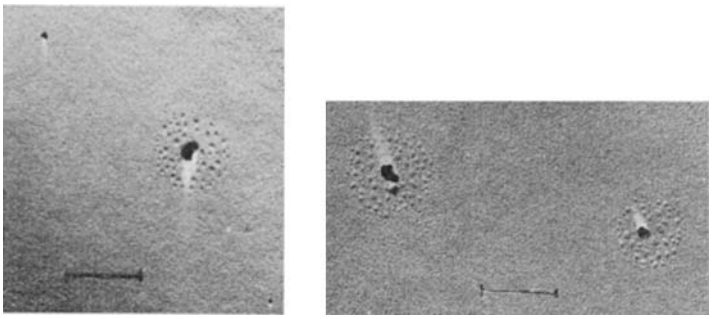

Fig. 1. Electron micrograph of sub-micron size solid particles collected from noctilucent clouds.

an ice coating on the particles prior to their collection. It must be emphasized that this interpretation has been inferred from the results of one single experiment and should therefore be treated with care. The collection experiment was repeated in 1964 with an improved instrumentation permitting altitude discrimination of the particles. Unfortunately, the results from the analysis carried out so far, which indicate an extremely low concentration of collected particles in both the cloud and the no-cloud flights, are inconclusive and do not corroborate the 1962 findings. It is thus still an open question if the formation of a light-scattering layer near the mesopause is to be ascribed to an increased number of concentration of solid particles caused by convergence or a time-dependent influx of extraterrestrial matter, to a local increase of the scattering cross-section due to ice sublimation, or to a combination of these mechanisms. In the present contribution the various aspects of mechanical accumulation will be disregarded and the discussion will be devoted to the problem of possibility and character of a sublimation process in the mesopause environment, and the changes of the optical characteristics of aerosol distributions produced by such a process.

Sublimation and evaporation at the mesopause

In order that a sublimation process be initiated and maintained in the presence of suitable nuclei, the actual water vapour mixing ratio must exceed the saturation mixing ration over ice at the temperature of the particles. A further condition is, however, that the supply of water molecules is not exhausted by the phase transition. The latter condition maximizes the particle size attainable by sublimation. The possibility of sublimation crucially depends upon the temperature distribution in the mesosphere. In order to establish the mesospheric temperature profile during the season of occurrence of noctilucent clouds series of rocket-grenade temperature soundings were conducted during 1963 and 1964 from the Kronogård launching site by the Institute of Meteorology in co-operation with NASA. These soundings were carried out in the presence of noctilucent clouds and also when no clouds were observed. The results from the Kronogård soundings, which yielded significantly lower mesopause temperature values than previously recorded at lower latitudes, are summarized in Figure 2. The experimental points which refer to the mean temperature in layers of varying geometrical thickness as indiciated by vertical lines, are denoted by different symbols for different launching dates. The presence of visually observed noctilucent clouds is marked by N. The magnitude of the estimated experimental error is indicated by horizontal lines. A theoretically derived temperature profile due to Nicolet (1960) and the AFCRL Standard Atmosphere profiles are also shown for comparison. It should be noticed that a low mesopause temperature is not necessarily associated with the appearing of a noctilucent cloud display; temperature values close to 130°K have been recorded in the presence as well as in the absence of the clouds. These results are corroborated by similar findings from recent rocket-grenade soundings at Point Barrow (71° N) and in the U.S.A. (Smith, 1965).

Regarding the abundance of the water molecule in the mesosphere our knowledge is scarce. Measurements of the vertical distribution of

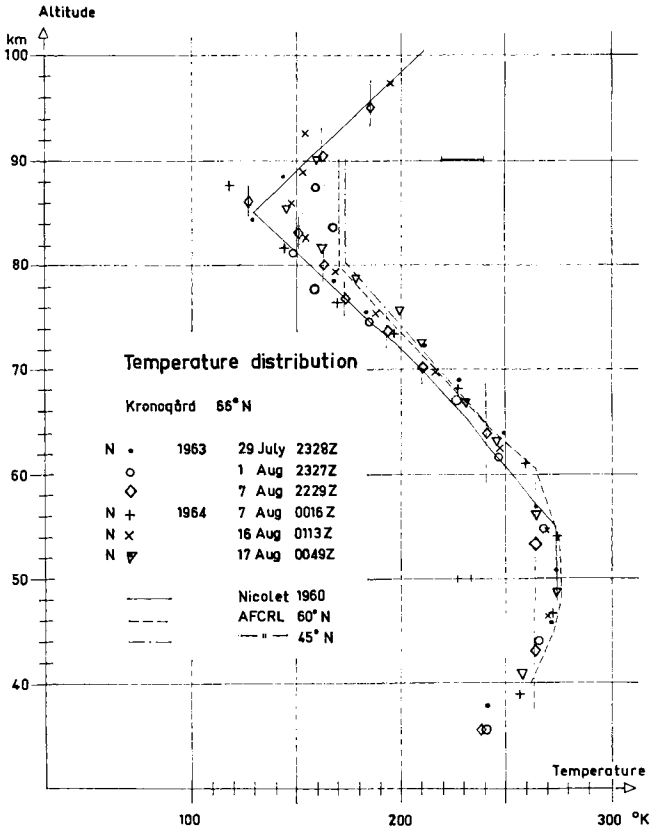

Fig. 2. Temperature as function of altitude from rocket-grenade soundings over Kronogård (66° N) in Sweden. The presence of visible noctilucent clouds is indicated in front of the launching date by *N*. The estimated error at 50 km and 90 km altitudes is shown by horizontal lines. Notice that the symbols denote the average temperature in layers of varying geometric thickness as indicated by short vertical lines.

water vapour have been made up to 30–40 km altitude yielding widely diverging results. Some of these measurements indicate not only very high water vapour concentrations but also an increase of the mixing ratio with altitude. Using an improved measuring technique and careful analysis of his data Mastenbrook (1963) has been able to show that a large part of the water vapour found by earlier investigators has probably been carried into the stratosphere by the measuring equipment itself. According to Mastenbrook, the water vapour concentration in the upper stratosphere is not expected to exceed 10^{-6} to 10^{-5} g/g. Similar results were also reported by Goldsmith (1964).

In the mesosphere the water molecule is dissociated by solar ultraviolet radiation at wavelengths below 1900 Å. According to theoretical considerations by Bates & Nicolet (1950) this dissociation could be significant down to the 65 km level. In a static atmosphere photodissociation would result in a rapid decrease of the vapour concentration above 70 km. In recent theoretical studies of the photochemistry of hydrogen compounds in the high atmosphere Hesstvedt (1964, 1965) has shown that in the presence of a net upward transport in the mesosphere, the water molecule may be replaced at a faster rate than destroyed by photolysis. As a result the dissociation level is lifted and the equilibrium concentration of water vapour above 70 km increased. This effect is illustrated in Figures 2*a* and 2*b* by the broken lines marked *A* and *B*. These curves show the vertical profile of water vapour density derived from Hesstvedt's calculations which were carried out

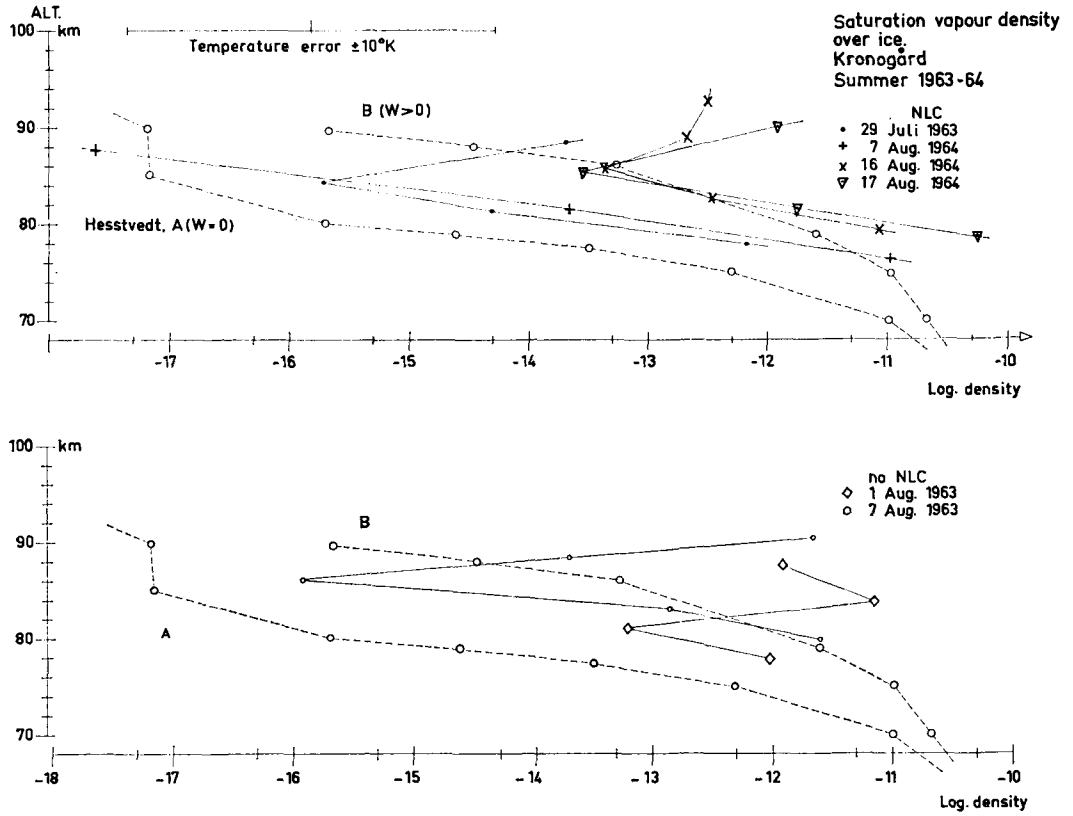

Fig. 3. (*a*) Saturation vapour density over a plane ice surface as function of altitude. The symbols indicate values derived from the results of the Kronogård rocket-grenade soundings, when noctilucent clouds were observed. Different symbols refer to different launching dates. The error in the saturation density that would be caused by a temperature error of $\pm 10°K$ is also indicated. The dashed curves show theoretically calculated vapour density profiles, (A) in a static atmosphere, (B) including the effect of upward transport (Hesstvedt, 1964, 1965). (*b*) Same as (*a*), though in the absence of noctilucent clouds. The saturation density was computed from an extrapolated formula (Smithsonian Met. Tables, 1963).

with the assumption of a very high stratospheric mixing ratio (1.5×10^{-4} g/g). Curves *A* refer to the case of a static atmosphere and curve *B* is based upon the circulation model of Murgatroyd & Singleton (1961). The various symbols in these figures denote values of the saturation vapour density over ice derived from temperature date from the Kronogård soundings; Figure 3*a* refers to cases when noctilucent clouds were present and Figure 3*b* when no clouds were observed. The magnitude of the error in the saturation vapour density associated with an error in the temperature of $\pm 10°K$ is indicated in Figure 3*a*.

With due regard to the gross simplifications and errors involved, it can be concluded that in a static atmosphere a sublimation process at the mesopause level is unlikely to occur, in particular if the more recent humidity data are adopted. In the presence of upward transport by mixing or by large-scale ascent, and at the low temperature prevailing at the high-latitude mesopause during summer, sublimation of ice would be possible.

In the rarified-gas environment in the mesosphere the dimensions of particles considered here are small compared to the molecular mean free path and the conditions of molecular free flow apply. The particle is assumed to be spherical and sedimenting at its terminal fall velocity. The rate of accretion of ice is determined by the frequency of encounters with Maxwell-distributed water molecules experienced by the

particle along its trajectory. As to the structural form of ice deposited at a slow rate in the temperature regime considered, little information is available. In the region 190 to 130°K experimental investigations have indicated a cubic structure; at lower temperatures the ice is supposed to be amorphous (Blackman & Lisgarten, 1958). For the present purpose it is assumed that the vapour molecules which come to rest on the surface will form a coating of amorphous ice. Throughout the following discussion the nucleus and coating are regarded as concentric spheres.

The growth rate of the ice coating is given by

$$dr/dt = \frac{\alpha_c}{2\varrho_i}\left\{\sqrt{\frac{2R_vT}{\pi}}\varrho_v - \sqrt{\frac{2R_vT_p}{\pi}}\varrho_s\right\}. \quad (1)$$

Here r denotes the particle radius, ϱ_v the ambient water vapour density and ϱ_s is the saturation vapour density over ice at the temperature of the particle, T_p. The ambient temperature is T and R_v is the gas constant for water vapour. The specific density of ice is ϱ_i and α_c is the condensation coefficient ($\alpha_c \leq 1$). For an ice surface this quantity is close to unity and throughout this discussion it is assumed that $\alpha_c = 1$. The saturation vapour density over a spherical particle is given by Kelvin's equation,

$$\varrho_s = \varrho_{so} \exp\left(2\sigma_{iv}/\varrho_i R_v T_p r\right), \quad (2)$$

where ϱ_{so} is the saturation vapour density over a plane surface and σ_{iv} the specific free energy of the ice-vapour interface.

The temperature of the particles is not necessarily equal to the ambient temperature, being determined by the balance between molecular heat transfer and the absorption and emission of radiation. Due to the slow sublimation rate expected during mesopause conditions the effect of latent heat release is negligible. It can be shown that owing to a high absorptivity in the visible, and poor emissive power in the infrared spectrum, sunlit particles composed by an electrically conducting substance such as e.g. iron are not suitable sublimation nuclei in the mesosphere. Dielectric particles do not absorb visible radiation and the radiative energy exchange of such particles is confined to infrared spectral regions. Compared with molecular heat transfer the latter is negligible throughout the mesosphere. The temperature of dielectric particles is therefore always close to that of the ambient air. The radiative heat balance of sub-micron particles, and the dependence of absorption and emission upon the particle size and refractive index has been discussed in detail by Van de Hulst (1946, 1949).

The radius-dependence of the saturation vapour pressure sets a further limit to the range of suitable sublimation nuclei. To obtain a rough estimate of the magnitude of the curvature correction, assume that the value of $\sigma_{iv} = 100$ erg/cm^2 can be applied at the presently considered low temperatures (Dufour & Defay, 1963). With $R_v = 4.615$ erg/g/deg, $\varrho_i = 0.91$ g/cm^3 and $T = 140$°K, the correction factor is 1.46 for $r = 10^{-6}$ cm and 30.5 if $r = 10^{-7}$ cm.[1] The possibility of the existence of regions at the mesopause level where the degree of supersaturation is extremely high cannot a priori be excluded. Even in this case, however, particles with radii smaller than about 0.01 microns are expected to grow only in the absence of competitive nuclei in their environment. The possibility of initiation and maintenance of the sublimation process and the features produced by such a process depend on both the composition, the number concentration, and the form of size distribution of solid particles at the mesopause. For particles with radii greater than about 0.01 microns, to which the subsequent discussion is confined, the effect of curvature is small as long as supersaturation prevails. The rate of change of the coating thickness in the supersaturated environment is practically independent of the radius. The highest possible deposition rate can be estimated by assuming that within the supersaturated layer $\varrho_v \gg \varrho_s$ and that the second term in the parenthesis in (1) is negligible. Inserting numerical values into the growth equation it is found that at a temperature of 140°K and with a vapour density of 10^{-13} g/cm^3 corresponding to a mixing ratio of about 10^{-5} g/g at the 80 km level, the growth rate can not exceed 6×10^{-8} cm/min. With $\varrho_v = 10^{-14}$ g/cm^3 this value is ten times smaller. As the available supply of water molecules is impoverished by the phase transition and downward transport by sedimentation, the maximum speed can only be maintained during the initial phase of the sublimation process. By similar argumentation it can be shown that unless the temperature is drastically

[1] Neglecting the effects of possible electrical charges on the equilibrium vapour pressure as well as the possibility of hydrated ions as potential sublimation nuclei.

changed, the evaporation process at the mesopause is slow. Particles formed by sublimation can exist for a considerable time even in a presence of a vanishingly small vapour concentration. At a temperature of 150°K the saturation vapour density would be 8.77×10^{-14} g/cm³. If ϱ_v is neglected in comparison with ϱ_s the maximum rate of evaporation is still only 5.6×10^{-8} cm/min. On this basis it is possible to define a characteristic time scale for optical changes by sublimation and re-evaporation in noctilucent clouds. At the horizontal translation velocities observed in noctilucent clouds the time scale of optical changes is small compared with that of horizontal displacements. This limits the value of inferences obtained from local temperature measurements as to the question whether sublimation is indeed involved with the formation of the light-scattering layer.

The final coating thickness attainable by a sedimenting particle depends upon the length of time it spends in the supersaturated layer. This time is determined by the vertical extent of this layer and the fall-speed of the particles. In molecular-flow conditions the settling velocity relative to the air is proportional to the radius and the specific density of the particle. For a 0.1 micron-particle at the mesopause this speed is of the order of 10 cm/sec. A further important parameter is, of course, the vertical velocity of the air. The length of time available for the accretion of ice varies inversely with nucleus size and therefore the final coating thickness is expected to be greater on the smaller nuclei. As to the relative increase of the particle radius by the coating the smaller particles are even more favoured. If the number concentration of nuclei increases with decreasing radius then the sublimation process will eventually result in a steepening of the slope of the size distribution function. This effect, which is of principal importance to the optical response of an aerosol to a sublimation process, has been demonstrated by Charlson (1966), who has formulated a simple, steady-state model of noctilucent clouds and integrated numerically the growth equation under various assumptions regarding the ambient water vapour concentration and speed of air ascent.

Consider once more the evaporation process. While the rate of coating deposition was limited by the available concentration of water molecules, the limit to the speed of evaporation is solely set by the local temperature. A temperature rise of 10°K changes the saturation density by a factor geater than 10. Thus the evaporation process may under circumstances be very rapid. Below the mesopause the observed temperature gradient is about 5 to 7°K and because of the small fall velocities involved, evaporation will take place in a relatively shallow layer once the particle leaves the saturated environment. Therefore, if sublimation is a necessary condition for the formation of a visible cloud layer, rapid evaporation at the cloud base is sufficient to account for the observed distinct boundary. It should be pointed out that since the water vapour released by evaporation remains in the neighbourhood of the mesopause, sedimentation in the ice phase is expected to lead to redistribution of the water molecule in the mesosphere. In the presence of ascending air these molecules will be available for further sublimation, which obviously also applies to particles whose fall-speed is smaller than the updraught velocity.

Optical properties of aerosol layers

General considerations

The specific intensity of the scattered light observed in the presence of a noctilucent cloud display is

$$I = I_b + I_c, \tag{3}$$

where I_b is the twilight background illumination and I_c represents the total contribution to scattering by the aerosol layer. The two components of the latter with the electrical vectors vibrating perpendicularly and parallel to the scattering plane as indicated by the subscripts $k = 1$ and 2 are expressed by

$$I_k = F_0 q_1 q_2 \tau P_k(\vartheta)/8\pi \cos i. \tag{4}$$

Here q_1 is the selective atmospheric attenuation of the illuminating solar flux F_0 and q_2 is the attenuation of the scattered light. The normal optical thickness is τ while P_k is the appropriate component of the average scattering phase matrix for scattering angle ϑ. The P_k depend upon the form of the vertically averaged size distribution function and τ depends in addition upon the total number concentration of the aerosol particles. The geometrical factor $(\cos i)^{-1}$ represents the changes in the apparent optical thickness of the cloud layer due to the varying angle i between the normal to the layer and the direction of vision.

The effect of variable atmospheric transmission and background illumination upon the variability of noctilucent cloud sightings will not be discussed here. The importance of the geometric factor in connection with the classification of noctilucent clouds according to their "brightness" needs, however, to be emphasized. In the presence of undulations in the cloud surface the angle i will undergo local variations which will give rise to substantial brightness variations, particularly pronounced at low elevation angles. In the vicinity of very deep waves the geometric enhancement of the scattered intensity may be as high as a factor 10, which under circumstances may be necessary to permit visual observation. Measurements of the contrast of noctilucent cloud features indicate that even in intense displays the intrinsic brightness of the strongest features remains below $10^{-5}\ F_0$. The estimated normal "optical thickness", or more precisely the ratio I/F_0 between the specific scattered intensity and the illuminating flux at wavelengths and scattering angles where these clouds are observed, is of the order of 10^{-6} or less.

The exact shape of the particles in noctilucent clouds, and the vertical variation of their concentration and size distribution have so far not been directly determined. For the present discussion it will be sufficient to consider the case of independent, single scattering by homogeneous distributions of hypothetical, spherical particles. The particles are assumed to have uniform refractive index which is a real number for dielectric, and complex for absorbing particles. On the basis of the previously discussed character of the sublimation process the coated particles will be treated as concentric spheres, the refractive index of the outer shell being that of ice, i.e. $m = 1.31$.

The intensities of the two perpendicularly polarized components of the light scattered by a spherical particle are proportional to the scattering functions $i_1(x, mx, \vartheta)$ and $i_2(x, mx, \vartheta)$ where the dimensionless size parameter is $x = 2\pi r/\lambda$ for a wavelength λ and particle radius r, m is the refractive index, and ϑ is the scattering angle. For a homogeneous sphere the scattering functions are obtained from the Mie theory (Van de Hulst, 1957). For concentric spheres a modified form of the Mie theory is available (Güttler, 1952; Aden & Kerker, 1952). In the latter case the intensity functions depend on the individual size parameters and refractive indices of the nucleus and the coating. The polarization of the scattered light is given by

$$P = (i_1 - i_2)/(i_1 + i_2) \tag{5}$$

and the colour ratio may be defined as the ratio between the monochromatic scattering function for a given scattering angle at different wavelengths

$$C = [i_1 + i_2]_{\lambda a}/[i_1 + i_2]_{\lambda b} \quad \text{where} \quad \lambda_a > \lambda_b. \tag{6}$$

Because of their independence of the absolute number concentration the latter parameters, if properly corrected for background illumination, are particularly convenient for aerosol identification.

On the basis of the radius dependence of the scattering properties the size spectrum of small particles may be divided into three different size ranges. The scattering by very small particles ($x \ll 1$) is Rayleigh scattering characterized by a symmetric scattering pattern and throughout positive polarization which is almost complete at $\vartheta = 90°$. Within this size range

$$i_1 = x^6 |\mu|^2, \qquad i_2 = I_1 \cos^2 \vartheta, \tag{7}$$

where $\mu = (m^2 - 1)/(m^2 + 2)$. The scattered intensity is proportional to r^6 and $C = (\lambda_a/\lambda_b)^{-6}$. The size range of large particles ($x > 5$) is characterized by a highly anisotropic scattering pattern with a pronounced forward peak. The polarization produced by such particles is small and oscillates between positive and negative values except in the case of absorbing particles. Between these size regions there is an interval of gradual transition from Rayleigh to Mie scattering. This size range contains the particles here defined as "optically efficient" by the fact that such particles would yield the greatest amount of scattered light obtainable from an ensemble of particles with given total mass. Characteristic for this size region is the gradually increasing preponderance of forward scattering increase of the colour index (reddening) and, for dielectric particles, a decrease of polarization with increasing radius. For brevity particles belonging to the above defined size classes will be called Rayleigh, Mie, and efficient particles.

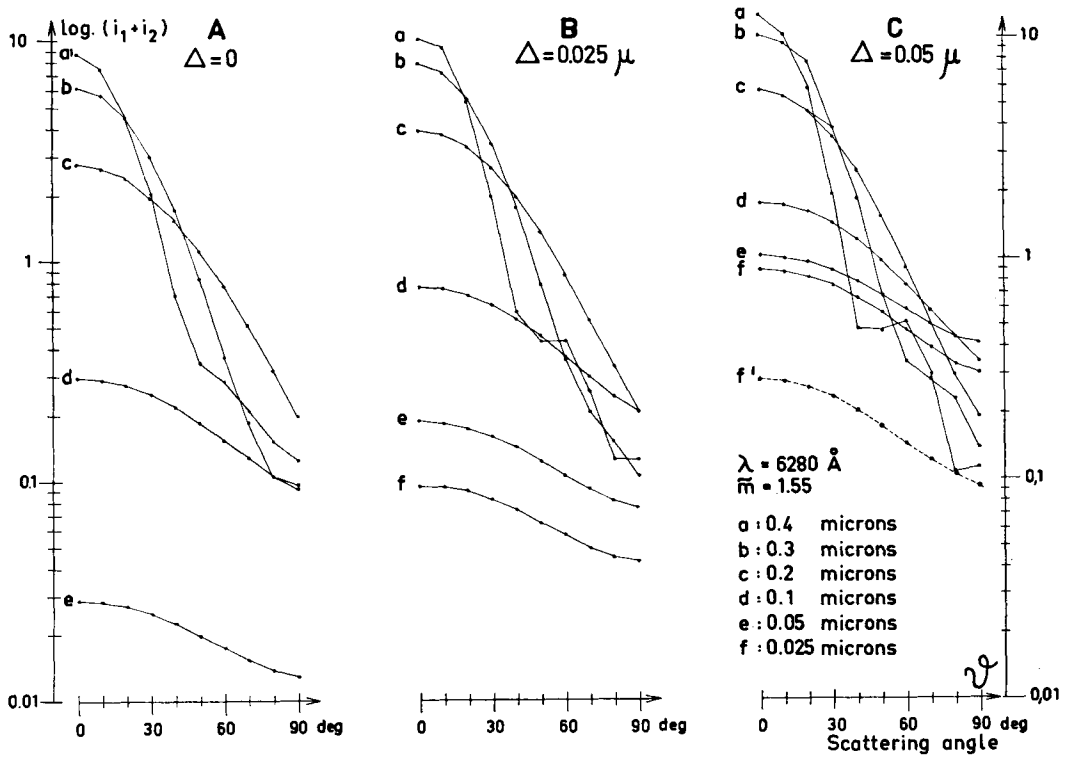

Fig. 4. Scattering patterns of uniform-size distributions of dielectric spheres ($m = 1.55$). Diagram A refers to uncoated particles and diagrams B and C to the same particles coated by an ice shell of 0.025 and 0.05 micron thickness. Different curves refer to different nucleus radii, as indicated. The vertical logarithmic scale is the scattering function of a volume containing a given total mass of particulate matter = the mass of one particle with $r = 0.1$ microns. The total particle number in each distribution is $(r/0.1)^{-3}$ if r is in microns. The wavelength is 6280 Å. Curve f' in diagram C would be obtained if only 3 of the 32 particles in distribution f are coated and the coating thickness is 0.075 microns.

Scattering by coated particles

The variations in the scattering characteristics following the deposition of dielectric coating depend on the size range to which the nucleus and the composite particle belong, and also on the refractive index of the nucleus. Within the Rayleigh region the magnitude of the scattering functions increases rapidly with the radius, while the angular dependence of intensity, polarization and colour ratio are unaltered. If the coated particle enters the transition region the intensity increase becomes gradually reduced, the scattering pattern increasingly anisotropic and the process is followed by reddening. The polarization is reduced and attains eventually negative values at certain scattering angles.

In order to study the optical properties of aerosol distributions and the possible optical effects of sublimation, the scattering functions of concentric two-layer spherical particles were numerically computed. The complete Mie solution was used throughout the calculations and recurrence relations were used to obtain the transcendental functions required. Integrals were evaluated by Simpson's formula. Results from the calculations will be used below to describe some of the essential optical features of the presently considered sublimation process. It should be observed that the aim is not to formulate an optical model of noctilucent clouds which with the evidence available is not yet possible.

The case of a monodisperse aerosol is considered first. Examples of computed scattering functions together with the changes upon the deposition of an ice coating are shown in

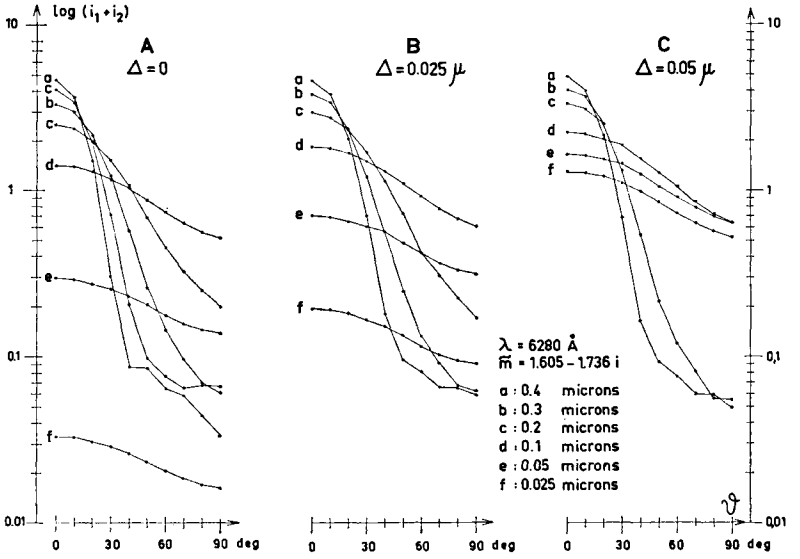

Fig. 5. Same as Fig. 4, but for metallic (iron) nuclei.

Figure 4. The curves refer to a uniform-size distribution of nuclei where the number concentration is inversely proportional to the cube of the radius. Each function corresponds to the same total amount of solid matter, namely that of one particle with $r = 0.1$ microns (distribution *d*). The wavelength is 6280 Å, the refractive index of the nucleus is $m = 1.55$ and that of the ice coating $m = 1.31$. The appropriate values of r are indicated. Diagram A refers to the uncoated particles, diagrams B and C to the coated particles with a coating thickness of 0.025 microns (B) and 0.05 microns (C). The marked increase of the scattering function of Rayleigh particles upon coating deposition (curves *e* and *f*), which should be noticed, is accentuated by the increasing ratio between the final and the initial radius with decreasing particle size. Analogous features are exhibited by similar diagrams in Figure 5 which apply to metallic (iron) nuclei. Owing to the complex refractive index, the scattering by metallic Rayleigh particles is comparatively strong, as seen from curves *d*, *e*, and *f*, diagram 5 A.

From the point of view of noctilucent clouds it is important to consider that the total amount of water vapour necessary to produce a given brightness gain varies with the size of the nuclei. Curve *d* in diagram *C* of Figure 5, for example, refers to a single particle which carries a total mass of ice of approximately 10^{-14} g, while the amount required to produce curve *f* is about ten times larger. If, however, only 3 of the 64 particles of distribution *f* are allowed to obtain a coating, then curve *f'* would be obtained at the expense of only 1.2×10^{-14} g of ice. For comparison, the highest estimated water vapour concentration above the 80 km level is 10^{-14} to 10^{-13} g/cm³. In the presence of high concentration of very small nuclei with equal probability to be coated, the available supply of water vapour might beexhausted by the sublimation process without producing observable optical effects. It is emphasized that when the above discussion is applied to actual conditions at the mesopause, the time available for coating deposition, which depends on both the particle size, the updraught velocity and the thickness of the super-saturated layer, must be considered. Assume, for example, that sublimation is possible in a layer of 1 km thickness and that the average growth-rate throughout the layer is 2×10^{-9} cm/min. Then, the time required to deposit a 0.02 micron thickness coating is 10^3 min, and a particle whose fall-speed is greater than 1 m/min would traverse the layer before this coating thickness can be attained unless the fall-speed is reduced by the air ascent.

Theoretical circulation models generally de-

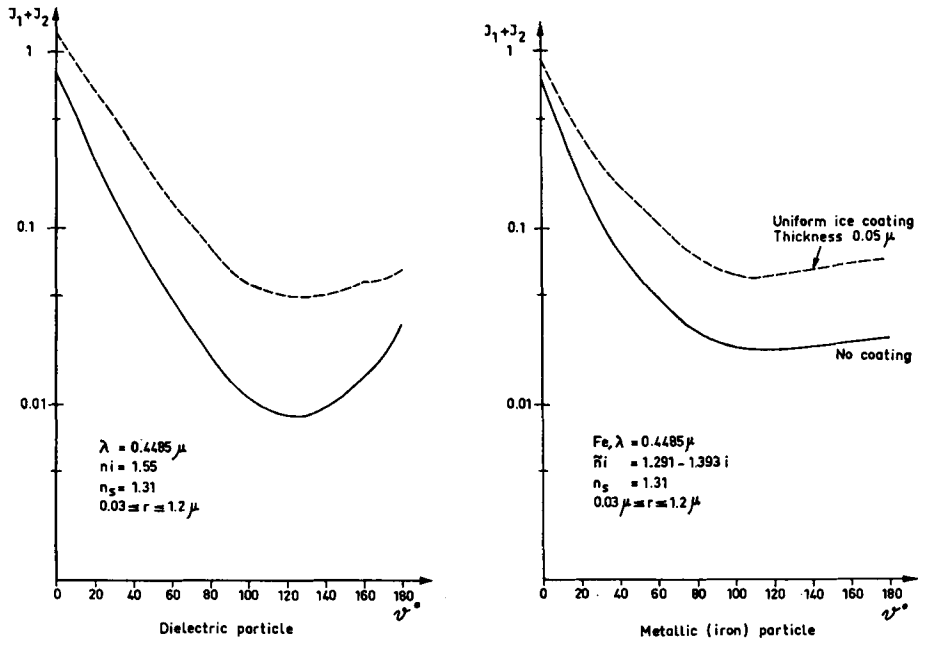

Fig. 6. (*a*) Average scattering pattern of a model distribution of dielectric spheres obeying the distribution function $dn/dr \sim r^{-4}$, $0.03 \leqslant r \leqslant 1.2$ microns. The solid line refers to uncoated particles, the broken curve to the same aerosol but with each particle coated by an ice shell of 0.05 microns thickness. The curves are normalized with respect to the particle number and the absolute scattering function for N particles with $r > r_0$ microns is obtained by multiplying the vertical scale by $3N$ $(r_0/0.05)^3$. Notice the reduced preponderance of forward scattering following the coating process. (*b*) Same as (*a*), but for absorbing (iron) nuclei for comparison.

scribe the mean state of the mesosphere and the actual vertical velocities may locally be substantially greater than those derived from model calculations. Where this occurs, it is possible that ice sublimation on Rayleigh particles could alone produce the number of optically efficient particles required to create a distinct scattering layer.

Polydisperse particle distributions

The scattering functions of a mixture of homogeneous particles which obey a size distribution law $dn/dr = f(r)$ are given by

$$I_k(\vartheta) = \int_{r\min}^{r\max} i_k(\vartheta) \cdot f(r)\, dr \quad (k = 1, 2). \qquad (8)$$

The same formula applies to coated spheres with due observance of the possible dependence of the coating thickness upon the nucleus size. It should be observed that in nature this relationship may vary from point to point within the aerosol layer.

The integrand in (8) possesses in most cases a definite maximum corresponding to a different radius at different scattering angles and wavelengths. If the size distribution can be approximated by a power law, as is often the case in the atmosphere, then at scattering angles beyond about 20° the maximum contribution to the integrand originates from the "optically efficient" particles. The optical properties of the aerosol are determined by the abundance of particles within this size region, which also defines the "mean" particle size derived from photometric measurements. The changes produced by a radius modification process such as sublimation depend upon the change of concentration of particles within this region. The relative increase of the particle radius by the presently discussed sublimation process is greater the smaller the particle size. By virtue of the higher concentration of the smaller particles the coating deposition would give rise to an increased dominance of the characteristic features of small-particle scattering, at least at

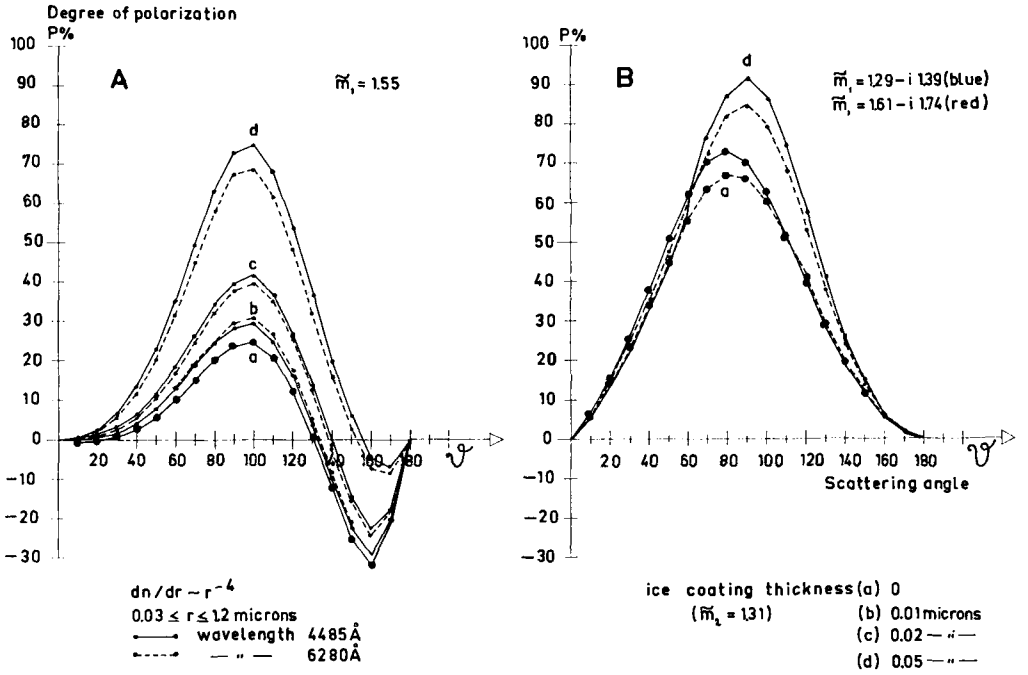

Fig. 7. (*a*) Degree of polarization as function of the scattering angle corresponding to the same aerosol distribution as in Fig. 6*a*. The solid lines refer to $\lambda = 4485$ Å, the broken lines to $\lambda = 6280$ Å. The coating thickness is different for different pairs of curves, as indicated. Notice the characteristic negative polarization in the backward hemisphere and the increase of polarization upon the coating deposition. (*b*) Same as (*a*), but for iron nuclei.

scattering angles beyond 20°. Thus the brightness enhancement would be accompanied by an increase of the polarization, and decrease of the colour ratio, or bluening, which is most pronounced at large scattering angles. A further consequence of the coating process is the reduction of the preponderance of forward scattering. An obvious pre-requisite for these changes to occur is that the aerosol contains an adequate supply of nuclei smaller than the "efficient" particles, i.e. smaller than $r \sim 0.1$ microns for visible wavelengths. The above outlined features are illustrated by a set of graphs which show the computed scattering parameters, and the changes in these parameters upon the deposition of an ice coating referring to a power-law distributed hypothetical aerosol of spherical particles. The coating is assumed to be independent of the radius. The power-law distribution function was selected for the present purpose by the observational evidence that unless noctilucent clouds are formed by an isodisperse aerosol, the number concentration of particles must decrease with increasing radius throughout the range of optically efficient particles. In view of the previously discussed character of the sublimation process, the choice of the radius-independent coating might be questioned. Yet, by the fact that the presently discussed features would only be further accentuated if the coating thickness is greater on the smaller particles, this assumption is sufficient for its purpose.

Figures 6 to 8 show in turn the average scattering pattern, the polarization, and the colour ratio as functions of the scattering angle. The size distribution is $dn/dr = Ar^{-4}$, known also as the Junge distribution, r ranging from 0.03 to 1.2 microns. The thickness of the ice coating is 0.05 microns independently of the nucleus size. Figure 6*a* refers to dielectric particles ($m = 1.55$) and for comparison, Figure 6*b* to iron nuclei at $\lambda = 4485$ Å. The solid lines refer to uncoated, the broken lines to the coated particles. The scattering patterns of both kinds of nuclei are characterized by the pronounced forward peak, due to the presence of large particles. This pre-

ponderance of forward scattering is less for a distribution function with a steeper slope, and greater for lower refractive index. The ratio of scattered intensities at 20° and 100° scattering angles for the uncoated particles in Figure 6*a* is thus 22.0, and for an r^{-5} distribution would be 13.6; if $m = 1.4$ the corresponding numbers are 37.0 and 19.3. With the exception of a narrow angular region in the forward and backward directions, the scattered light by dielectric nuclei is almost entirely contributed by particles with radii between 0.075 and 0.4 microns. The indicated values of the scattering function are normalized with respect to the particle number. To obtain the corresponding functions for a scattering volume containing N particles with radius greater than r_0, the vertical scale is to be multiplied by a factor $3N \cdot (r_0/0.05)^3$ if r_0 is in microns.

Figures 7*a* and *b* show the polarization as function of the scattering angle for two wavelengths, namely 4485 Å (solid lines) and 6280 Å (broken curves). The pronounced negative polarization in the backward region is characteristic for dielectric particles. The increase of the polarization with the coating thickness, and the shifting of the polarization maximum toward $\vartheta = 90°$ is observed even in the case of the strongly polarizing absorbing particles. The variation of the colour ratio with the scattering angle, and its changes upon the coating deposition are shown in Figure 8. For a power-law distribution with exponent p the parameter $C = (\lambda_a/\lambda_b)^{1-p}$ provided that the range of integration extends well beyond the boundaries of the size range of maximum contribution. This theoretical value is indicated by broken horizontal lines in the diagram. Deficiency of particles with radii within this size range, as well as a distortion of the slope of the distribution function (e.g. by a sublimation process), would give rise to deviations in the colour ratio at the proper scattering angles. Thus, for dielectric nuclei the decrease of C or bluening at forward and backward angles observed in the figure arises from the absence of particles larger than $r = 1.2$ microns, while the reddening exhibited by the metallic particles is due to a similar deficiency at the small-particle limit. The bluening caused by increasing dominance of the small-particle scattering characteristics which follow the coating deposition is common for both distributions. In view of the different behaviour

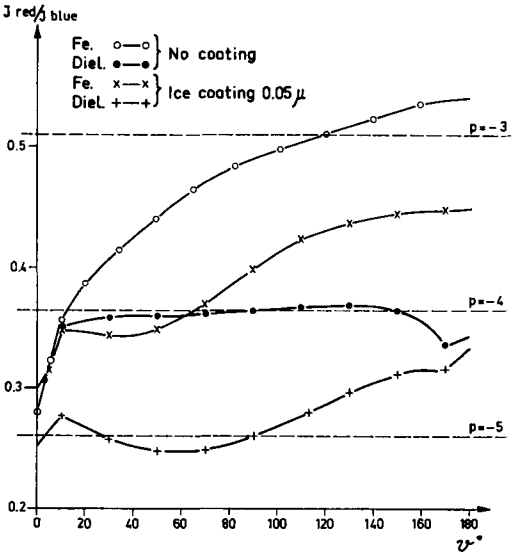

Fig. 8. Colour ratio as function of the scattering angle. The aerosol model is the same as in Fig. 6*a*. The dashed horizontal lines denote theoretical values corresponding to different values of the power-law exponent p. The curves refer to uncoated and coated dielectric and absorbing nuclei, as indicated. The model aerosol is as in Figs. 6*a* and *b*.

of the uniform-size, and the polydisperse aerosol, as outlined above, the interpretation of colour and polarization variations alone in terms of ice sublimation is difficult. The expected coating deposition rate is slow compared with horizontal wind speeds usually observed in the mesopause region and changes of the scattering characteristics by sublimation may in addition be blanketed by transport processes and spatial inhomogeneities in the aerosol distribution.

Figures 9*a* and *b* finally show the angular distribution of brightness enhancement by the coating process. The vertical scale is the ratio of intensities before and after the application of an ice coating with a thickness of 0.02 microns, and 0.04 microns as indicated. Each figure shows computed values referring to an r^{-4} and an r^{-5} distribution with $m = 1.4$ (Fig. 9*a*) and $m = 1.55$ (Fig. 9*b*). The efficiency of the uniform-thickness coating is as expected greatest for the steeper distribution function and for the lower refractive index. The strong increase of I/I_0 in the backward direction reflects the higher symmetry of the scattering pattern of the coated particles.

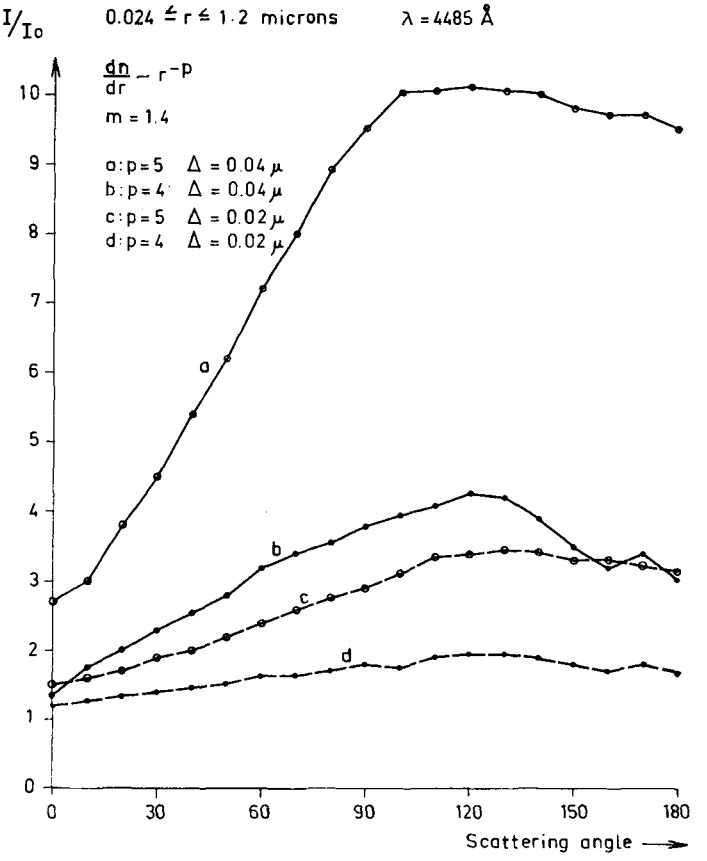

Fig. 9 (*a*).

Fig. 9. (*a*) Relative increase of the scattered intensity upon deposition of an ice coating on to dielectric ($m = 1.4$) particles which obey an r^{-p} distribution law. The different values of p, and coating thickness Δ are indicated. (*b*) Same as (*a*), but for nuclei of a higher refractive index ($m = 1.55$).

Assuming an average total number of one particle per cm³ the mass of ice per unit volume carried by a uniform coating thickness of 0.04 microns would be about 0.17×10^{-14} g/cm³ if $p = 4$, and 0.14×10^{-14} g/cm³ if $p = 5$. The total number per unit area of the coated particles which would be required to produce a scattering layer with $I/F_0 = 10^{-6}$ at $\lambda = 4485$ Å, and at scattering angles 20° and 60° is shown in Table 1, where the unit is $N \times 10^{-5}$/cm² column. The values in parentheses refer to the uncoated particles.

The occurrence of the above outlined features at visible wavelengths depends critically upon the character of the small-particle limit of the size distribution of possible sublimation nuclei. To investigate this limit future optical measurements should be extended to wavelength domains where the smallest particles beome "optically efficient", i.e. the ultraviolet spectrum.

Table 1

	$p = 4$		$p = 5$	
	$m = 1.55$	$m = 1.4$	$m = 1.55$	$m = 1.4$
20°	0.23	0.25	0.85	1.0
	(0.44)	(0.50)	(2.8)	(3.8)
60°	1.1	1.5	2.5	3.1
	(2.7)	(4.6)	(12.4)	(22.6)

From the assumption of the radius-independent coating thickness, the theoretical numbers given above represent a lower limit.

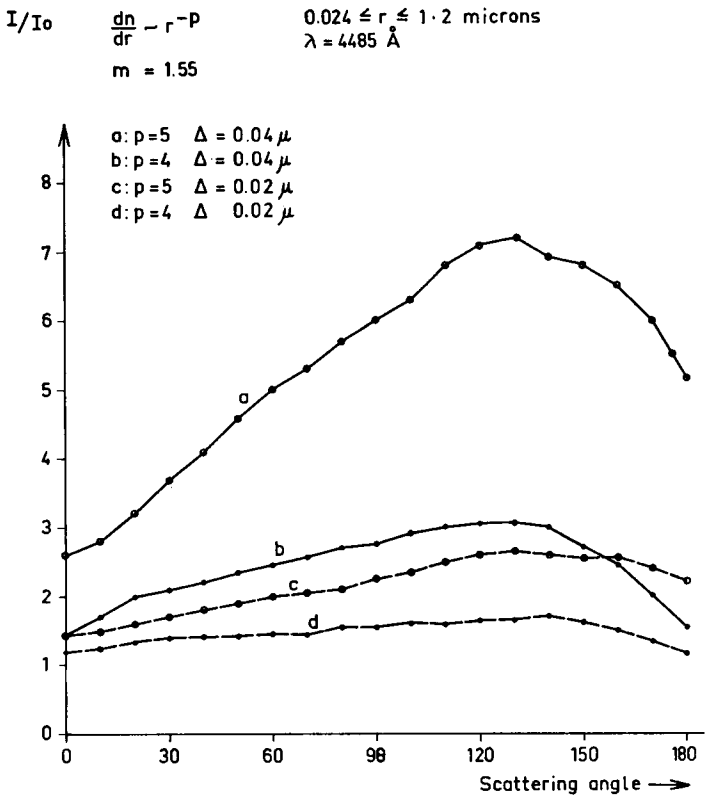

Fig. 9 (*b*).

Concluding remarks

From the preceding discussion it is evident that in order to formulate a realistic model of noctilucent clouds the size distribution as well as the concentration of possible sublimation nuclei must be carefully considered. The observed optical properties of noctilucent clouds allow the following conclusions:

1. The brightness and scattering characteristics of the clouds at scattering angles beyond 20° are determined by the relative abundance of particles in the size range defined approximately by $0.05 \leqslant r \leqslant 0.4$ microns. The formation of a layer with adequate optical thickness by Rayleigh particles would require improbable concentrations of these particles at the mesopause.
2. The role of ice sublimation in the formation of noctilucent clouds, and the modification of their optical properties by this process, depend critically on the presence and concentration of particles with radii smaller than about 0.1 microns, known as Aitken nuclei.
3. Provided that an adequate concentration of these particles is present in the mesopause region, the sublimation process alone may be sufficient and perhaps necessary to account for the formation of a distinct, strongly polarizing scattering layer. Depending on the nucleus distribution, the coating deposition may be necessary to permit visual observations at large scattering angles, such as the observer's zenith. Available experimental data are not sufficient to resolve this question and further information is required especially with regard to the presence of Aitken nuclei in the mesosphere in no-cloud conditions.
4. If the characteristic time-scale of the sublimation process is defined by the time required to change the brightness by a factor 2, then this time is at least 10 to 100 minutes.

5. Below the mesopause region the evaporation rate increases rapidly with decreasing altitude and it is fast in comparison with the settling velocity of sub-micron particles. This process might be sufficient to account for the distinct boundary of the aerosol layer. Further conclusions regarding this question require information from continued height-dependent particle sampling experiments. Such experiments are presently being prepared at the Institute of Meteorology (Wilhelm & Witt, 1966).

To conclude this contribution, various possible optical methods expected to provide information regarding the nature of mesospheric aerosol particles might be summarized. The importance of determining each relevant parameter by more than one independent procedure should be emphasized since the incomplete knowledge of these parameters may lead to ambiguous interpretatation of the experimental results. It is thus important that future rocket-borne particle collection experiments (such as currently planned by the Institute of Meteorology) should include *in situ* measurement of radiance and polarization of the scattered light, preferably in different regions of the spectrum. Such measurements will furnish information about the altitude of the scattering layer as well as about the optical properties of the aerosol particles. The merits of ground-based photometry of noctilucent clouds and of the twilight sky will not be discussed here. The possibility of obtaining useful information by investigation of the day-to-day variation of the Babinet and Arago neutral points during the summer twilight may, however, be worth mentioning.

Ground-based optical measurements are influenced by the turbidity and by single and multiple Rayleigh scattering in the lower atmosphere. This difficulty can be overcome by rocket, and also balloon-borne photopolarimetry, which offers the additional advantage of extending these measurements to short-wave ultraviolet wavelengths where substantial deviations from the Rayleigh scattering law may be caused by very small particles which scatter light according to this law in the visible spectrum. For height discrimination of various scattering layers, and for the study of the presence and possible changes of the aerosol layer in the absence of sunlight, the pulsed-laser backscatter technique is a promising tool. Unfortunately, this method involves formidable difficulties and no decisive results have hitherto been reported (Fiocco, 1966). Further laser backscatter experiments are being conducted at the Institute of Meteorology, and elsewhere.

The last experiment to be mentioned here is the photometric detection of light scattering layers in the mesosphere from a space vehicle. Regarding twilight conditions similar experiments have previously been proposed by Deirmendjian (1963), and a satellite borne observation of noctilucent clouds has also been discussed by Rosenberg (1965). The problem is, however, to investigate the possible occurrence of noctilucent clouds in the sunlit portion of the atmosphere, where illumination conditions preclude visual observation. Knowledge of the daytime distribution, and the presence in polar regions of the clouds, would be essential to any theory about the mechanism of formation and maintenance of these clouds. The technique suggested is to utilize spectral regions where the illumination of the lower-lying atmosphere is strongly reduced by selective absorption, and where the presence of solid particles is expected to cause polarization anomalies or an increase of the atmospheric albedo. Such regions are e.g. the Hartley band of ozone at 2500 Å to 2700 Å wavelength, and the Schumann-Runge bands of molecular oxygen ($\lambda < 1950$ Å). An experiment of this kind would also be feasible from rockets. In fact, preliminary rocket-borne photopolarimetric investigation of noctilucent clouds in the 2550 Å-region is on the current experimental programme of the Institute of Meteorology. A satellite-borne instrument would, on the other hand, scan the atmosphere continuously, independently of weather conditions, for a long period of time, thus being an essential supplement to a world-wide network of ground-based observers.

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REFERENCES

Aden, A. L. & Kerker, M. 1952. Scattering of electromagnetic waves from two concentric spheres. *J. Appl. Physics 22*, 10, 1242–1246.

Bates, D. R. & Nicolet, M. 1950. The photochemistry of atmospheric water vapor. *J. Geophys. Res. 55*, 301–327.

Blackman, M. & Lisgarten, N. D. 1958. Electron diffraction investigations into the cubic and other structural forms of ice. *Advances in Physics 7*, No. 26 (April 1958).

Charlson, R. J. 1966. A simple noctilucent cloud model. Proceedings of the CACR Symposium, Visby 1965. *Tellus 18* (2–3), 451–456.

Deirmendjian, D. & Vestine, E. H. 1958. Some remarks on the nature and origin of noctilucent cloud particles. *Plan. Space Sci. 1*, 146–153.

Deirmendjian, D. 1963. Detection of mesospheric clouds from a satellite. In *Proceedings of the First Internat. Symposium on Rocket and Satellite Meteorology*, 406–411, North-Holland Publ. Co., Amsterdam.

Dufour, L. & Defay, R. 1963. Thermodynamics of Clouds. *International Geophysics Series 6*. Academic Press, New York and London, 1965, 103–106.

Fiocco, G. & Grams, G. 1966. Observations of the upper atmosphere by optical radar in Alaska and Sweden during the summer 1964. *Tellus 18*, 34–38.

Goldsmith, P. 1964. *Measurements of humidity up to 30 km using a new hygrometer*. Paper delivered at the International Atmospheric Ozone Symposium, Albuquerque, New Mexico, September 1964.

Güttler, A. 1952. Die Miesche Theorie der Beugung durch dielektrische Kugeln mit absorbierendem Kern und ihre Bedeutung für Probleme der interstellaren Materie und des atmosphärischen Aerosols. *Annalen der Physik 11*, (Heft 2–3), 65–89.

Hemenway, C. L., Soberman, R. K. & Witt, G. 1964. Investigations of noctilucent cloud particles. *Tellus 16* (1), 84–109.

Hesstvedt, E. 1964. On the water vapor content in the high atmosphere. *Geofysiske Publikasjoner 25* (3), 1–18.

Hesstvedt, E. 1965. On the spatial distribution of some hydrogen components in the mesosphere and lower thermosphere. *Tellus 17* (3), 341–349.

Mastenbrook, H. J. 1963. The status of water vapor observations above 20 km and implications as to the general circulation. Paper delivered at the Upper Atmosphere Meteorology Symposium, IUGG, Berkley, California, August 1963.

Murgatroyd, R. J. & Singleton, F. 1961. Possible meridional circulations in the stratosphere and mesosphere. *Quart. J. Roy. Met. Soc. 87*.

Nicolet, M. 1960. The properties and constitution of the upper atmosphere. In *Physics of the Upper Atmosphere*, Ed. Ratcliffe, 55, Academic Press, New York.

Rosenberg, G. V. 1965. On the observability of noctilucent clouds and nacreous clouds from a spacecraft. English Ed. of *Izvestiya AN SSSR, Atmospheric and Oceanic Physics 2* (1), 21–27.

Smith, W. S. 1965. Private communication. *Smithsonian Meteorological Tables*, 1963, 6th ed., 350.

van de Hulst, H. C. 1957. *Light Scattering by Small Particles*. John Wiley & Sons, New York.

van de Hulst, H. C. 1946, 1949. The solid particles in interstellar space. *Recherches Astronomiques, Obs. d'Utrecht 11*, parts 1–2.

Wilhelm, N. & Witt, G. 1966. Aspects concerning the design of a noctilucent cloud particle collecting experiment. Presented at the *International Symposium on Noctilucent Clouds*, Tallinn, March 1965.

Willmann, Ch. 1962. On the polarization of light from noctilucent clouds. *Trudi Soveschanya po Serebristym Oblakam*, Tallinn.

Witt, G. 1960. Polarization of light from noctilucent clouds. *J. of Geoph. Res. 65*, 925–933.

ОПТИЧЕСКИЕ ХАРАКТЕРИСТИКИ РАСПРЕДЕЛЕНИЯ АЭРОЗОЛЯ В СВЯЗИ С С ЕРЕБРИСТЫМИ ОБЛАКАМИ

До сих пор невозможно было установить, появляются ли серебристые облака вследствие увеличения концентрации, или вслед ствие локального увеличения размеров твердых частиц в мезопаузе, или вследствие совместного действия обоих этих механизмов. В данном сообщении на основе современных представлений о структуре и фотохимии мезосферы обсуждается связь между оптическими характеристиками серебристых облаков и сублимацией льда.

Первая часть посвящена обсуждению характера процесса сублимации на этих высотах. Показано, что такой процесс возможен в условиях, превалирующих в высокоширотной мезопаузе летом. Временной масштаб

сублимации и испарения является медленным по сравнению с горизонтальными скоростями переноса, наблюдаемыми в этой области. Ниже мезопаузы испарение становится быстрым по сравнению со скоростью переноса субмикронных частиц.

Во второй части на основе численных расчетов индикатрис рассеяния сферических частиц, покрытых слоем льда, дается обзор рассеивающих свойств изодисперсных и полидисперсных распределений частиц и возможных изменений, вызванных процессами сублимации. Найдено, что оптические последствия сублимации критически зависят от присутствия и относительной концентрации частиц, вызывающих рэлеевское рассеяние в видимом спектре. Характерный временной масштаб изменений яркости при сублимации составляет, по крайней мере, величину от 10 до 100 минут.

Суммируются различные оптические методы для дальнейшего изучения природы мезосферного аэрозоля и предлагается метод для определения рассеивающих слоев в мезосфере, использующий измерения интенсивности ультрафиолетовой радиации и поляризации с ракет и спутников.