Radiative Transfer Through a Cloud of Absorbing-Scattering Particles

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An experimental study was made of radiative transfer through a plane-parallel enclosure of micron-sized, spherical aluminim oxide particles and flake-like graphite particles. By suspending the particles in both carbon tetrachloride and carbon disulfide, transmission measurements were made at wave lengths from 2 to 11 microns with both collimated and diffuse incident radiation. *In correlating the diffuse results, the two-flux model was rearranged so that the cloud* transmission was expressed in terms of two cloud scattering parameters. The values of the scattering parameters as determined from the data were correlated with the properties of the cloud.

Radiant heat transfer involving media which both absorb and scatter the electromagnetic radiation is in general a most complex problem. Chandrasekar (*1*) gives a comprehensive discussion of the formulation of the equation of transfer and its solution under a variety of boundary conditions. Synoptic discussions are also included in several recent texts on radiative transfer (*7, 13, 21*). As alternatives to the rigorous solutions of the equation of transfer (including numerical techniques) several approximate solutions have also been suggsted. Of these, the two-flux model first suggested by Schuster (*20*), and Kubelka and Munk (*11*), and extended by Hamaker (*6*), and Churchill and Chu (*3*), can be utilized without the availability of a digital computer. Even so, the relationships appear to be quite involved.

The application of any of the models, rigorous or approximate, requires a knowledge of the interaction between the individual particles and the electromagnetic wave. These interactions, the amount of radiation scattered, and the relative intensity of radiation in each direction depend upon the size, shape, and orientation of the particles and the optical properties of the particles and the surrounding medium. Furthermore, the total interaction between the cloud and the radiation depends upon the concentration of particles in the cloud and the distribution of particle sizes. The interaction between single particles and electromagnetic radiation has been treated extensively (*10, 15, 23*). The determination of the scattering properties in a cloud of particles is computationally complex even when the particles are assumed to act independently. This investigation adapts the two-flux model for radiative transfer through a plane-parallel cloud so that a solution can be described by parameters related to the properties of the cloud, without the intermediate determination of the scattering properties of the individual particles and the calculation of the combined effect of all particles.

Theoretical Model

The two-flux model divides the diffuse radiation field in two fluxes, one forward and one backward. The model has been utilized by others (*2, 6, 12*). Tien and Churchill (*22*) compared it to other approximations as well as to the exact solution of the transfer equation. One of the faults of the two-flux model is that it does not give an accurate representation when the incident radiation on the cloud is not axially symmetric. For such cases the six-flux method of Churchill and Chu (*3*) appears more suitable. However, for axially symmetric incident radiation the six-flux model reduces essentially to the same two differential equations as the two-flux model. In this investigation diffuse incident radiation on a plane parallel isothermal cloud was considered and the incident radiation was axially symmetric and the two-flux model was appropriate.

An energy balance on both the forward and backward fluxes leads to the following differential equations. For the forward flux

$$-\frac{dI_+(\tau)}{d\tau} = (1-\omega_0 f)I_+(\tau) - \omega_0 bI(\tau) - (1-\omega_0)I_B(\tau) \quad (1)$$

The first term on the right accounts for energy from the forward flux scattered in the forward direction. The second term accounts for energy from the backward flux scattered in the forward direction. The last term accounts for energy emitted by the particles. The equation for the backward flux is similar:

$$\frac{dI_-(\tau)}{d\tau} = (1-\omega_0 f)I_-(\tau) - \omega_0 bI_+(\tau) + (1-\omega_0)I_B(\tau) \quad (2)$$

The optical depth τ is defined by the equation

$$d\tau = K_t\left(\frac{L_e}{L}\right)\, dz \quad (3)$$

At low temperatures the emission by the cloud can be considered negligible.

For a plane-parallel cloud of thickness L with no reflectances at the cloud boundaries the boundary conditions are

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$$@ \quad z < 0, \quad I_+(0) = I_0 \tag{4}$$

$$@ \quad z > L, \quad I_-(L) = 0 \tag{5}$$

For the boundary conditions given, the solution for the transmission through a cloud is given by

$$\frac{I_+(L)}{I_0} = \frac{2}{(1-\psi)e^{-\sigma_d vL} + (1+\psi)e^{\sigma_d vL}} \tag{6}$$

The reflectance by the cloud is given by

$$\frac{I_-(0)}{I_0} = \frac{\sqrt{\psi^2 - 1}\,(e^{\sigma_d vL} - e^{-\sigma_d vL})}{(1+\psi)e^{\sigma_d vL} + (1-\psi)e^{-\sigma_d vL}} \tag{7}$$

The multiple scattering parameters, σ_d and ψ are defined by

$$\sigma_d = \sqrt{(1 - f\omega_0)^2 - (b\omega_0)^2}\ (L_e/L)\sigma_t \tag{8}$$

and

$$\psi = \frac{1 - f\omega_0}{\sqrt{(1 - f\omega_0)^2 - (b\omega_0)^2}} \tag{9}$$

A value of 1.0 is customarily assigned to the ratio (L_e/L), the ratio of effective depth to actual depth. Hottel and Sarofim (7) suggest that the true three-dimensional motion of the photon could be better represented by a single component if the value of (L_e/L) were 1.76. However, the proper value of the ratio should depend upon the angular distribution of the scattered energy within the cloud. If the ratio (L_e/L) is considered as an unknown it is not possible to determine experimentally the proper values of f, b, ω_0 and L_e/L. Nagy and Lenoir (17) determined f, b, and ω_0 for aluminum oxide particles suspended in carbon tetrachloride and carbon disulfide by assigning L_e/L a value of 1.0. Each of the parameters, σ_d and ψ, is defined in terms of quantities which are functions of the optical properties of the cloud. It should be possible to develop empirical correlations for the multiple scattering parameters σ_d and ψ.

The parameters defined above have physical significance which can be demonstrated by examining limiting cases of the solution. For example, for the optically thick limit the change in transmission with change of the product (vL) is given by

$$\left[\frac{d \ln \dfrac{I_+(L)}{I_0}}{d(vL)}\right]_{L\to\infty} = -\sigma_d \tag{10}$$

The above equation is directly analogous to the equation defining the extinction coefficient for particles exposed to collimated incident radiation. The analogy suggests that σ_d can be considered as a diffuse extinction coefficient. The significance of the parameter ψ can be illustrated by considering the total reflectance of an optically thick cloud. For a semi-infinite cloud with diffuse incident radiation the reflected relative intensity is given by

$$\left(\frac{I_-(0)}{I_0}\right)_{L\to\infty} = \sqrt{\frac{\psi - 1}{\psi + 1}} \tag{11}$$

which shows that the parameter ψ is a measure of the back scattering by the cloud. Figure 1 shows how the different values of ψ affect the transmission through a plane-parallel cloud. At large optical depths the slope of the transmission curve is relatively independent of ψ. Large values of the parameter ψ result in greater curvature of the transmission curve at small optical depths. In fact it can be shown that the initial slope of the curve is dependent upon both ψ and σ_d. In the optically thin limit the change of transmitted intensity with optical depth is given by

$$\left[\frac{d \ln \dfrac{I_+(L)}{I_0}}{d(vL)}\right]_{vL\to 0} = -\psi\sigma_d \tag{12}$$

Assuming the accuracy of the two-flux model Figure 2 shows the calculated values of $I_+(L)/I_0$ as a function of the amount of forward scattering and of the albedo ω_0, the fraction of extinction which is scattered. For an albedo of zero, corresponding to absorption with no scattering, the value of the parameter ψ is 1.0 regardless of the value of f, the fraction of radiation scattered in the forward direction. As the value of the albedo is increased, the value of ψ increases, approaching infinity as the albedo approaches 1.0, everywhere except at the singular point $f = 1.0$. It is worth noting that for even moderate amounts of absorption, for example 10%, the maximum value of ψ is 2.0. The effect of different values of σ_d and ψ on the transmission through a plane-parallel cloud and the importance of the scattering properties of the cloud in determining the values of σ_d and ψ have been given here to aid in under-

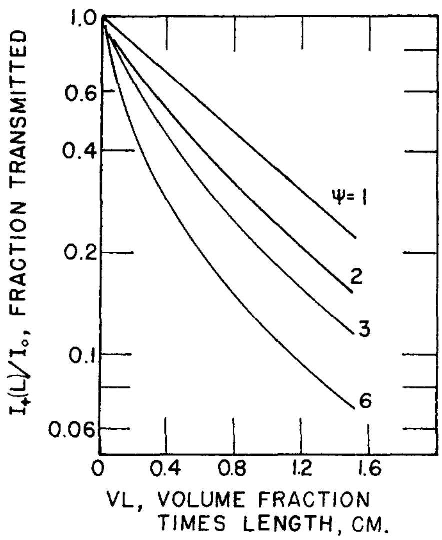

Fig. 1. The effect of the parameter ψ on the transmission through a plane-parallel cloud of diffuse incident radiation, $\sigma_d = 1.0$.

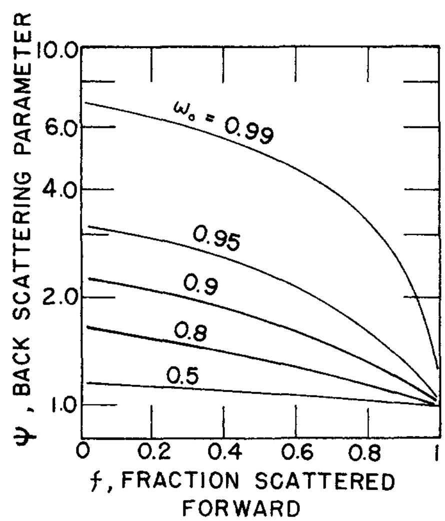

Fig. 2. The effect of forward scattering on the parameter ψ.

standing the interpretation of the experimental data and some of the associated problems.

EXPERIMENTAL PROCEDURE

The experimental procedure used in this investigation was essentially that reported by Nagy (*16*) and summarized by Nagy and Lenoir (*17*), the difference being in sample cells and sample handling procedures (*19*). A Beckman IR-2A Spectrometer was used to measure the transmission through clouds of aluminum oxide particles and graphite particles each suspended in each of two liquids, carbon tetrachloride, and carbon disulfide. Two different sizes of aluminum oxide particles were used, one with a volume average diameter of 16.1 microns and the other with a volume average diameter of 20.2 microns. The size distribution was determined by the use of an optical microscope. For the smaller size, 91% of the particles were included between the diameters of 13 and 19 microns. For the larger, 93% of the articles were included by 19 and 24 microns. In general the particles appeared to be near perfect spheres, but about 1 in 20 was irregular in shape.

The graphite particles were irregularly shaped flakes of natural graphite. Three different sizes of graphite particles were used with average Feret diameters* of 2.94, 5.47 and 10.3 microns. The diameters refer to the facial diameters of the particles. The statistical significance of the average Feret diameter has been considered by Walton (*24*) and Gebelein (*5*). The thickness of the flakes was calculated based on the reported surface area of the particles. The diameter to thickness ratio of the particles was 36. The distribution of diameters were such that for the three samples, respectively, 90% were between 1.6 and 4.8 microns, 93% between 1.6 and 10 microns, and 90% between 3 and 19 microns.

The particle clouds were prepared by mixing a stock suspension of each particle sample in one of the liquids. The suspension was well agitated to obtain good dispersion and a portion of the suspension dispensed into the sample cell. Additional clear liquid was then added to the cell and the ratio of suspension into liquid was varied to vary the concentration of particles in the cloud. The concentration of particles in each measured sample was determined by dumping the entire contents of the cell and weighing the contents before and after evaporation of the liquid.

The sample cell was made from a block of aluminum and had a sample cavity consisting of a right circular cylinder 2.38 cm. in diameter and 0.762 cm. long, when the cell was assembled. Two different window materials were used; Servofrax (arsenic tri-sulfide) and Irtran-3 (Kodak). The Irtran windows were used when carbon tetrachloride was the suspending liquid. The lower refractive index of the Irtran-3 reduced errors due to internal reflections.

Both collimated and diffuse transmission measurements were made for the 10 combinations of particle samples and liquids described before. For the diffuse source the front window of the sample cell was replaced by a Servofrax window with one surface roughened by No. 80 grit paper. This was the same window used by Nagy (*16*). Nagy examined the effectiveness of the roughened window as a diffuser. He also considered the effects of forward scatter on the transmitted beam from a collimated source. Nagy and Lenoir (*17*) reported that the forward scatter was not significant. Transmission measurements were made for both collimated and diffuse sources over a range of wavelengths from 2 to 11 microns.

* Feret diameter is the orthographic projection of particle cross section onto a fixed axis of reference.

TABLE 1. REFRACTIVE INDICES OF PARTICLES

Wave length, microns	Aluminum oxide	Graphite
2	1.738	1.906-*i* 1.037
3	1.712	2.167-*i* 1.295
4	1.675	2.399-*i* 1.486
5	1.624	2.598-*i* 1.635
6	1.535	2.770-*i* 1.760
8	1.344	3.047-*i* 1.972
9	1.210	3.163-*i* 2.070
10	1.084	3.268-*i* 2.164
11	1.000	3.364-*i* 2.257

EXPERIMENTAL DATA

The transmission measurements obtained from the infrared spectrometer were plotted on semilog coordinate paper. For the cases with collimated incident radiation the plots of transmission against volume fraction solids in the cloud fell along a straight line that could be easily drawn through the data. The extinction coefficients for each diameter and wave length were obtained from the slopes of the lines in accordance with the equation

$$\frac{d \ln \frac{I}{I_0}}{d(vL)} = -\sigma_t \tag{13}$$

which defines the particle extinction coefficient. The extinction efficiencies were determined by dividing the extinction coefficients by the geometrical cross sections of the particles.

For the cases using a diffuse source the data did not fall along a straight line. The values reported for σ_d and ψ are the best least square values corresponding to the equation

$$\frac{I_+(L)}{I_0} = \frac{2}{[(1 - R_0R_L) - (1 + R_0R_L)\psi + (R_0 + R_L)\sqrt{\psi^2 + 1}]e^{-\sigma_d(v + \alpha/\sigma_t)L} + [(1 - R_0R_L) + (1 + R_0R_L)\psi - (R_0 + R_L(\sqrt{\psi^2 + 1}]e^{\sigma_d(v + \alpha/\sigma_t)L}} \tag{14}$$

which is a modification of Equation (6) necessary to account for the absorption of the liquid and the reflectances of the cell windows at the cloud boundaries.

In applying Equation (14) the reflectance of the diffuse window was taken as zero, and the values of R_L for the plane windows were calculated hemispherical average values assuming equal incident intensity at all angles and no absorption within the windows. The scattering parameters determined were not sensitive to the values of R_0 and R_L used.

The extinction efficiencies for the collimated tests were correlated by the phase lag method of Van de Hulst (*23*). The phase lag in radians is equal to $(2\pi/\lambda)$ times the optical path difference between a light ray which passes through the particle along the diameter and one parallel to it in the suspending medium. For a spherical particle the phase lag can be represented by

$$a = \frac{2\pi d}{\lambda}(n_p - n_m) \tag{15}$$

When the two rays are in phase, they will reinforce each

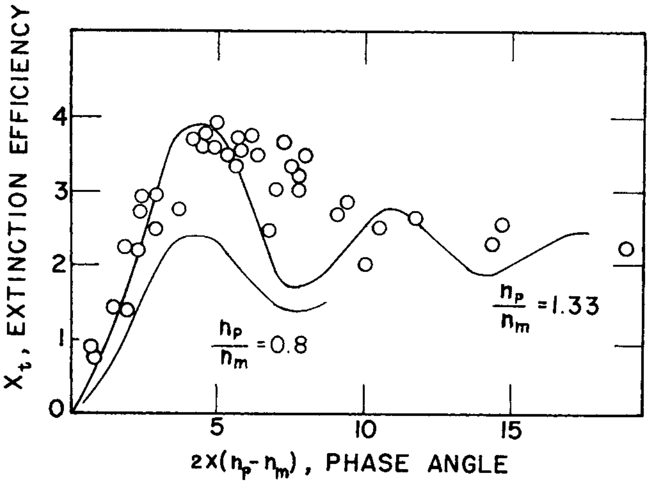

Fig. 3. Extinction efficiencies of aluminum oxide particles compared to theoretical values for uniform dielectric spheres.

other and the extinction efficiency will be lower. When they are out of phase the extinction efficiency will be higher.

Figure 3 shows the variation of the extinction efficiencies for aluminum oxide as a function of the phase angle based on average diameter. The scatter in the data is not unexpected since the relationship between the extinction efficiency and the phase angle is a function of the relative refractive index. For comparison, the relationship between the extinction efficiency and the phase angle for dielectric spheres with relative refractive indices of 0.8 to 1.33 as reported by Van de Hulst are also shown in the figure. There is reasonably good agreement between the absolute value of the phase angle at which the maximum value of the extinction efficiency occurs and the value expected from the theoretical curves. However, the data do not show a well defined minimum which would be expected at an absolute value of 7 or 8 for the phase angle. The succession of progressively smaller maxima and minima with increasing phase angle shown for the theoretical curves hold only for particles and suspending media which have a completely real refractive index. If absorption occurs either in the particle or the surrounding medium that phase of the cloud will have a complex refractive index and the relative refractive index will also be complex. Johnson, et al. (*8*) have shown that even a small value for the complex portion of the refractive index results in essentially complete disappearance of any maxima and minima beyond the first maximum observed. The distribution of particle sizes would also account for some smoothing of the maxima and minima. Furthermore, the aluminum oxide was crystalline which would also contribute to eliminating the maxima and minima.

The values of the refractive indices used for the calculations were those suggested and used by Nagy (*16*) which he based primarily on those reported by Malitson (*14*) for aluminum oxide and those reported by Pfund (*18*) and Kagarise (9) for the suspending liquids. The values of refractive indices of aluminum oxide for wave lengths from 6 to 11 microns were extrapolations. However, if the data for those wave lengths were excluded, the conclusions would not change. Actual differences between the refractive indices of the materials and those used in the calculations would account for some of the dispersion of the observed data. Treating the particles as one particle size while they actually had a finite size distribution would also contribute to some disagreement between the data and theoretical values.

One of the reasons for making the collimated tests was to determine the proper characterization of the particles. The parameters affecting the interaction of electromagnetic radiation and spherical particles is well treated in the theory. For randomly oriented particles Van de Hulst (*23*) showed that nonspherical particles might be considered as spherical if the proper equivalent diameter were chosen. For irregularly shaped particles the choice of an equivalent diameter is a prime consideration.

When the random orientation of the thin (diameter ≅ 36 × thickness) flake-like graphite particles is taken into account, the effective geometrical cross section of the particles is equal to ½ the facial area of the particles. This agrees well with Van de Hulst's (*23*) statement that the average geometrical cross section of a convex particle with random orientation is ¼ its surface area. Similarly, the effective thickness of the particle for use in calculating the phase angle is closely approximated by twice the thickness of the flake. The effective geometrical cross sections and thicknesses of the particles were used in calculating the extinction efficiencies and phase angles shown in Figure 4. The graphite particles were not dielectric and it was necessary to consider their complex refractive index. The dispersion equation of Dalzell and Sarofim (*4*) was used for calculating the refractive indices of the graphite. The extinction efficiencies of such particles depend on both the real and imaginary parts of the relative refractive index. Van de Hulst suggests that the extinction efficiencies be correlated against the phase angle and the quantity $\tan\beta$ where $\tan\beta$ is defined by

$$\tan\beta = \frac{n_p\kappa}{|n_p - n_m|} \tag{16}$$

For comparison with the data theoretical values of the extinction efficiencies at small phase angles based on Van de Hulst's approximation for complex refractive indices near unity are shown for three different values of $\tan\beta$: 1.0, 2.0, and 3.0. The data include values of $\tan\beta$ from 1.224 to 3.23 with the bulk of values falling between 1.3 and 1.5.

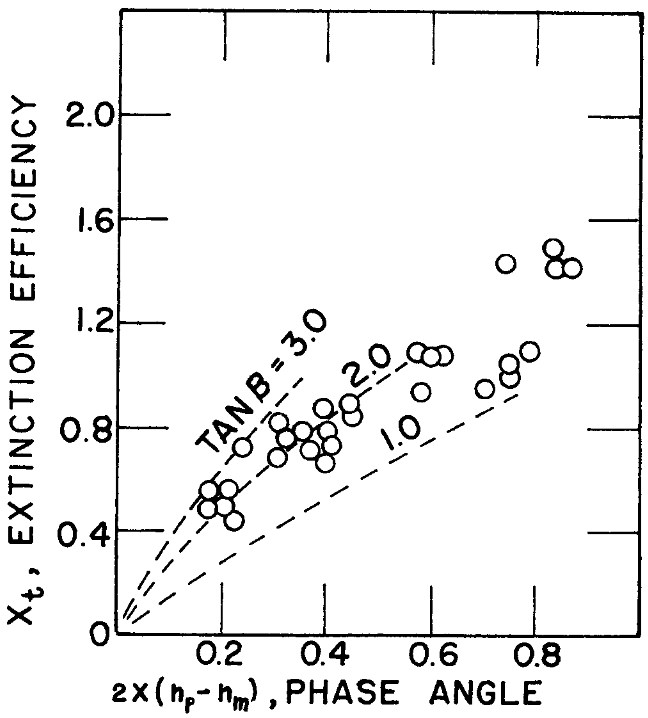

Fig. 4. Extinction efficiencies of graphite particles with collimated incident radiation.

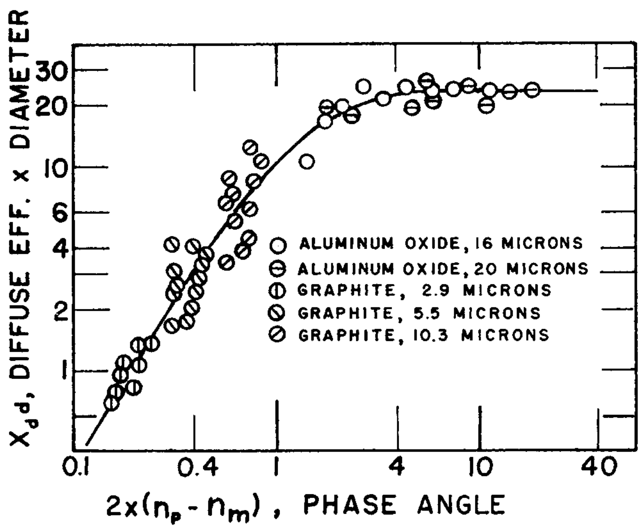

Fig. 5. "Diffuse" extinction efficiencies for positive phase angles.

It would appear from Figure 4 that the choices of effective geometrical cross section and effective thickness of the particle were reasonably suitable. While no direct comparison can be made between the data and the curves, the curves should approximate the data. If particle diameter were used to compute the phase angle the values would be 18 times greater and clearly unsuitable.

According to Equation (8), the diffuse extinction coefficient σ_d should be related to the total extinction coefficient σ_t. A correlation was found for all of the data of this study and could be represented by the equation

$$\sigma_d = 0.55\ \sigma_t \tag{17}$$

If the total extinction coefficient σ_t for the particles were known, or could be estimated, Equation (17) might be used to estimate a value for σ_d.

The nature of the diffuse extinction coefficient suggests defining a diffuse extinction efficiency by dividing σ_d by the geometrical cross section of the particles. Figure 5 shows a relationship between the diffuse extinction efficiency X_d and the phase angle. The curve shown in Figure 5 is given by the empirical equation

$$X_d d = 23.3(1 - e^{-a})^{1.82}, \quad a \geqq 0 \tag{18}$$

and holds only for positive values of the phase angle. For Equation (18) the average particle diameter d is expressed in microns. There appears to be a distinct difference between the magnitude of the diffuse extinction efficiency obtained with phase angles of the same magnitude but opposite in sign. This is not surprising when one considers that the diffuse extinction efficiency should be related to the hemispherical reflectance which is not the same on both sides of the interface between two substances. There was only a limited amount of data with negative values of the phase angle. They were for the aluminum oxide particles at wave lengths of 8 microns and above. Errors in extrapolating the refractive indices would not account for the higher extinction efficiencies. If it were necessary to estimate a value for the diffuse extinction efficiency with a negative phase angle the best estimate at this time would be to multiply the value obtained from Equation (18) by a factor of 2.6.

Figure 6 shows a plot of the back scattering parameter ψ as a function of the absolute value of the difference between the refractive index of the particles and that of the suspending liquid. The values of ψ shown in the figure are the optimum values of ψ determined from the transmission data when the values of σ_d are determined by Equation (18). There is appreciable scatter shown in the figure, reflecting the fact that the back-scattering of the cloud is not accurately determined by transmission data, except for optically thin clouds. Fortunately, it follows that predicted transmissions of plane-parallel clouds [by Equation (6)] are not sensitive to the value used for the parameter ψ.

The value of both of the parameters σ_d and ψ are affected by scattering and absorption, and analysis of the data as done in this study does not yield any direct measure of absorption or scattering by the particles. Some qualitative interpretations based on the values of ψ may be made with reference to Figure 2. Except when the fraction scattered forward is near 1.0 the values of ψ are not sensitive to the distribution of the scattered radiation. There the values of ψ are a rough measure of the albedo ω_0.

Actually values of ψ ranging from near unity to much larger values were obtained for both types of particles, as shown in Figure 6. The values of ψ near unity would indicate lower values of the albedo and therefore a greater portion of absorption relative to scattering. The shape of the curve in Figure 6 can also be explained somewhat through consideration of absorption versus scattering. In general larger differences between the refractive indices of the particles and the surrounding medium should mean that more of the incident radiation would be reflected at the particle surface and higher values of the albedo might be expected. However, Van de Hulst (*23*) has shown that when the difference is small and the particles have a relatively small absorption index the change in wave length may have a stronger effect on the albedo than a change in the refractive index difference. With the aluminum oxide particles increasing values of refractive index difference correspond to shorter wave lengths. For relative constant values of the absorption index more absorption within the particles would be expected. The same considerations will not explain the behavior of the values of ψ for the graphite particles, with which increasing refractive index difference corresponds to longer wave lengths. It would not be surprising if the data for the values of the parameter of ψ were best represented by a family of curves with the absorption index κ and the size parameter x affecting the values of ψ.

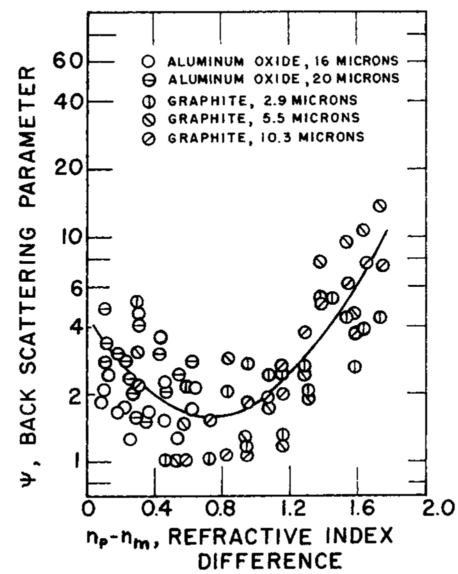

Fig. 6. The parameter ψ as a function of the difference in the refractive indices of the particles and the liquid.

The values of ψ are the most difficult to obtain with precision through transmission measurements. Precise values of ψ depend on precise measurements of transmission especially for low concentrations of particles in the cloud. In this study the precision of the transmission at low concentrations was limited by the problems of measuring the concentration itself. At high concentrations of solids it was difficult to measure precisely the low levels of transmission when a diffuse source was used. However, sufficiently precise measurements at moderate levels of transmission were obtained to yield reliable values for the parameter σ_d which is most important for the application of Equation (6).

To apply Equation (6) as suggested by the approach developed here depends on predicting the diffuse extinction coefficient σ_d through Equation (18). While the correlation represented by the equation was the best found for the data available, the manner in which the particle diameter d enters into the correlation is not understood. As the correlation stands, unreasonably large values of the diffuse extinction efficiency might be predicted for very small particles. Application of the correlation should probably be limited to particle sizes near the range included within the data of this study. While the correlation is very dependent at small phase angles on the data for graphite particles, the refractive indices used for the graphite do not appear to be critical, although better values might reduce the amount of scatter. The largest source of difficulty with the graphite particles was probably that of accounting for the irregular shapes. However, the measurements made with a collimated source indicated that proper considerations were taken.

In summary, this study has introduced a different approach to making calculations of diffuse radiative transfer through plane-parallel particulate clouds. The calculation method depends on the values of two parameters which have been related empirically to the optical properties of the particles and the surrounding medium. While refinements or improvements in the correlations can be expected, the method as presented is an alternative to considerably more complex methods for engineering calculations.

NOTATION

a = phase angle = $2x(n_p - n_m)$
b = fraction of scattered radiation in hemisphere opposite to direction of flux
d = particle diameter, microns
f = fraction of scattered radiation in the forward direction
i = $\sqrt{-1}$
I = intensity, radiant energy flux density per unit solid angle of divergence
I_B = intensity of black-body radiation
I_0 = intensity on entry into system of interest
$K(K_a, K_s, K_t)$ = cloud extinction coefficient or cross section per unit volume of cloud (subscripts refer to absorption, scatter, total extinction)
L = path length
L_e = effective path length
n = real part of refractive index
n' = complex refractive index = $n(1 - i\kappa)$
R_L = reflectance of the cloud boundary at $x = L$
R_0 = reflectance of the cloud boundary at $x = 0$
t = thickness of particle
v = volume fraction solids
x = size parameter: $(\pi d/\lambda)$ for spheres, $(2\pi t/\lambda)$ for flakes
z = coordinate in the direction
$X(X_a, X_s, X_t)$ = Mie extinction efficiency (subscripts refer to absorption, scatter, and total extinction)
X_d = diffuse extinction efficiency
α = absorption index of liquid, or medium surrounding the particles
β = defined by $\tan\beta = n_p\kappa/|n_p - n_m|$
κ = absorption index $K_a\lambda/4\pi n$
λ = wave length, microns
σ = $(\sigma_a, \sigma_s, \sigma_t)$ = particle extinction coefficient or cross section per unit volume of particles (subscripts refer to absorption, scatter, and total extinction)
σ_d = particle diffuse extinction coefficient
τ = optical thickness
ψ = multiple scattering parameter
ω_0 = albedo for scatter, the ratio of scatter to total extinction coefficient $K_s/(K_s + K_a)$

Subscripts

a, s, t = absorption, scatter, and total extinction
m = measured in the medium
p = measured in the particle
$+, -$ = used with intensity, in a unidimensional system, to indicate flux in the direction of increasing or decreasing value of the principal coordinate

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