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MEASUREMENT OF AEROSOL CHARACTERISTICS
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THE INVESTIGATION OF ADVANCED REMOTE SENSING
TECHNIQUES FOR THE MEASUREMENT OF AEROSOL
CHARACTERISTICS.

By

Adarsh Deepak 
Principal Investigator

Jacob Becher 
Co-Investigator

Final Report
For the period September 1975 - December 1977

Prepared for the
National Aeronautics and Space Administration
Langley Research Center
Hampton, Virginia

Under
Research Grant NSG 1252
M.P. McCormick, Technical Monitor
Instrument Research Division

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P.O. Box 6369
Norfolk, Virginia 23508

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THE INVESTIGATION OF ADVANCED REMOTE SENSING TECHNIQUES FOR THE MEASUREMENT OF AEROSOL CHARACTERISTICS

By
Adarsh Deepak

INTRODUCTION

This is the final report on investigations performed under research grant NSG 1252. The overall aims of the research program proposed under this grant were to conduct investigations of advanced remote sensing techniques and inversion methods for the measurement of characteristics of aerosol and gaseous species in the atmosphere. Of particular interest were the physical and chemical properties of aerosols, such as their size distribution, number concentration, and complex refractive index, and the vertical distribution of these properties on a local as well as global scale. In this connection, the principal investigator, Dr. Adarsh Deepak, provided the necessary science support to the NASA/Langley Research Center (NASA/LaRC) technical monitor on numerous projects outlined below, which involved development of remote sensing techniques for monitoring of tropospheric aerosols as well as satellite monitoring of upper tropospheric and stratospheric aerosols, development of computer programs for solving multiple scattering and radiative transfer problems, as well as inversion/retrieval problems. A necessary aspect of these efforts was to develop models of aerosol properties.

The research investigations have been categorized under the following topics, which are listed in the order in which they will be discussed in this report:

I. The Solar Aureole Method for Determining Aerosol Characteristics

II. Participation in University of Arizona-Aerosol and Radiation Experiment (UA-ARE), May 1977

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1 Research Associate, Old Dominion University Research Foundation, P. O. Box 6369, Norfolk, Virginia 23508, and Research Associate Professor, Department of Physics and Geophysical Sciences, Old Dominion University, Norfolk, Virginia 23508.
III. Multispectral Solar Radiometry for Determining Aerosol Size Distributions

IV. Aerosol Physical Properties of the Stratosphere (APPS) Experiment (The Draper Laboratory AAPE Project)

V. Stratospheric Aerosol Photographic Experiment (SAPE)

VI. The UCLA Tropospheric Polarimeter and Multispectral Radiometer Project (The Polarimeter and the Multispectral Radiometer as Remote Probes of Aerosols)

VII. International Interactive Workshop on Inversion Methods in Atmospheric Remote Sounding, Williamsburg, VA, December 15-17, 1976

VIII. Modeling of Aerosol Properties

This final report briefly describes some of the salient features of the research investigations conducted under grant NSG 1252 for the period September 1975 to December 1977 and, in addition, identifies some important problems in which further research should be continued in the near future.
I. THE SOLAR AUREOLE METHOD FOR DETERMINING AEROSOL CHARACTERISTICS

INTRODUCTION

The solar aureole method has been successfully used in recent years to determine size distribution and concentration of atmospheric aerosols, particularly tropospheric aerosols. A long-range objective of the research performed in this method was eventually to develop satellite- or Shuttle-based remote sensing systems for monitoring atmospheric aerosols.

The solar aureole is the area of enhanced brightness closely surrounding the Sun's disk (within about 20 degrees) caused mostly by aerosol scattering of sunlight. Since the aerosols scatter predominantly in the forward direction, the contribution of atmospheric haze to sky radiance for angles close to the Sun is roughly $10^2$ to $10^3$ times the contribution by molecular scattering; this is illustrated in figure 1. It is to take advantage of this large signal range that a simple, portable, photographic solar aureole measurement (PSAM) technique was developed at the University of Florida in 1970 (ref. 1) and has since been used to diagnose the aerosol size-altitude distributions (refs. 2 to 5), by using the aureole radiance measurements along the almucantar. The almucantar is a conical scan of constant solar zenith angle with the local zenith as the axis. A solar aureole measurement program was subsequently initiated at NASA/Langley Research Center in 1974, and the photographic solar aureole isophote (PSAI) method (refs. 6 and 7) was developed. Isophotes are lines or curves of equal radiance. The PSAI method is an extension of the solar almucantar aureole radiance (SAAR) method. In the latter, the radiance measurements taken along the almucantar are used to infer the aerosol properties. In the PSAI method, in addition to making the almucantar measurements, one takes advantage of the fact, which emerged from our computer studies, that the shape of the solar aureole isophotes is sensitively dependent on the characteristics of the aerosol size distribution. Suggestions for the use of solar aureole measurements to determine aerosol properties had also been made earlier by Deirmendjian (refs. 8 and 9) and other researchers (refs. 10 to 14). Solar aureole measurements, taken with scanning photometers for determining aerosol properties, have also been made by Shaw (ref. 15) and Twitty et al. (ref. 16).
The following sections briefly describe the theory, photographic measurements, and results of the PSAI method. For details of some of the theoretical aspects discussed here, see references 1 and 17.

SIMPLIFYING ASSUMPTIONS

The forward-scattered radiance, being highly sensitive to \( n(r) \), is relatively insensitive to effects due to aerosol refractive index, polarization, and multiple scattering (MS), a fact that helps in simplifying the inversion problem. In this paper, only the single scattering (SS) theory treatment is considered, which should help in understanding the difficulties involved in the inversion of aerosol scattering measurements. Therefore, one makes the following reasonable simplifying assumptions:

1. Particles are spherical; hence Mie theory results can be used in computations.
2. The atmosphere is horizontally homogeneous and vertically inhomogeneous.
3. Absorption effects are ignored by selecting to work in spectral regions for which atmospheric absorption is nil.
4. The polarization effects are small for forward-scattered light and can be ignored.
5. For relatively clear days (visibility greater than 15 km), the MS effects at the forward-scattering angle are small compared with SS effects (ref. 4) and can be ignored.
6. An average value for the refractive index of all atmospheric aerosols is assumed for forward scattering.
7. The atmosphere is treated as plane-parallel; the spherical Earth effects, which become significant for zenith angles \( \phi \) larger than 75 degrees, are incorporated into the theory by the use of the generalized Chapman-type functions \( S(\phi) \) (refs. 4 and 18) in place of the secant functions.

The case of retrieval of size distribution from solar aureole data in the presence of multiple scattering has been discussed as part of another contract report (ref. 19) and, therefore, will not be discussed here.
SINGLE SCATTERING THEORY OF THE SOLAR AUREOLE

Figure 2 illustrates the geometry of the calculation. Shown are an acceptance cone $d\Omega$ originating at the detector and a solid angle $d\Omega'$ centered at the elemental scattering volume $dV$ at altitude $y$(km); $\phi_s$ and $\phi$ are the zenith angles of the Sun and the narrow view cone, and $\omega$ is the dihedral angle between the normals to the Sun zenith and view cone zenith planes intersecting at $dV$. The scattering angle $\psi$ is then given by the relation

$$\cos \psi = \cos \phi \cos \phi_s - \sin \phi \sin \phi_s \cos \omega$$

The element $dV$ is given by

$$dV = R^2 \ dQ S(\phi) \ dy$$

where the generalized Chapman-type functions (refs. 4 and 17)

$$S(\phi) = \sec \phi \quad (\text{for } \phi \leq 75 \text{ degrees})$$

We make measurements in regions of the sky for which $\phi \leq 75$ degrees and $\phi_s \leq 75$ degrees; then, $dV = R^2 d\Omega \ sec \phi \ dy$.

The optical depth defined by

$$\tau_j(\lambda, y) = \int_y^\infty \beta_j'(\lambda, y) \quad (j = M, P)$$

for a ray traversing the distance from the Sun to the air mass element $dV$ is given by

$$\tau_1 = \Sigma_j \tau_j(\lambda, y) \ sec \phi_s \quad (j = M, P)$$

and from the air mass to the detector by
\[ \tau_2 = \sum_j \left\{ \tau_j(\lambda,0) - \tau_j(\lambda,y) \right\} \sec \phi \] (6)

where M denotes air molecules; P, the particulates (aerosols); \( \lambda \), the wavelength; and \( \beta' \) the volume scattering coefficient (VSC) (km\(^{-1}\)) at altitude \( y \) for the jth constituent. In this paper, all quantities represented by \( \tau_p, \beta'_p, \beta_p, F'_p, F_p, P'_p \), and \( P_p \) are functions of \( \tilde{m} (= m' - im") \), the aerosol complex refractive index. The primes denote the \( y \)-dependence of the quantities. The terms \( \beta'_p \) and \( \beta'_M \) are defined as

\[ \beta'_p(\lambda,y) = \int_{r_2}^{r_1} \eta(r,y) \pi r^2 Q(x,\tilde{m}) dr \] (7)

where \( Q(x,\tilde{m}) \) is the efficiency factor (ref. 20), \( x = 2\pi r/\lambda \) is the particle size parameter, \( r_1 \) and \( r_2 \) are the minimum and maximum values of \( r \), and

\[ \beta'_M(\lambda,y) = \beta_M(\lambda) \rho_M(y) \] (8a)

where the VSC for molecules is

\[ \beta_M(\lambda) = \frac{8\pi^3(n^2 - 1)^2}{N\lambda^4} \left( \frac{4 + 3d}{4 - 3d} \right) \] (8b)

In equation (8b), \( N \) is the number of molecules (cm\(^{-3}\)); \( n \), the refractive index of the medium, \( d = 4\Delta/(1 - \Delta) \); and \( \Delta \) is the depolarization of scattered light at a scattering angle of 90 degrees for a linearly polarized incident radiation with its electric vector perpendicular to the scattering plane. For unpolarized incident light, \( \Delta \) is replaced by \( \xi = 2\Delta/(1 + \Delta) \). Then, the volume scattering function (VSF) for air molecules \((j = M)\) is

\[ F'_M(\psi,\lambda,y) = \beta_M(\lambda) P'_M(\psi) \rho_M(y), \quad \text{km}^{-1}\text{sr}^{-1} \] (9)

where the molecular phase function is

\[ P'_M(\psi) = \frac{3}{16\pi} (1 + \cos^2 \psi), \quad \text{sr}^{-1} \] (10)

6
where $\rho_M(y)$ is the dimensionless function representing the altitude distribution of molecular density. The VSF for aerosols ($j = P$) is

$$F^i_P(\psi, \lambda, y) = \beta^i_P(\lambda, y) P^i_P(\psi, \lambda, y)$$

(11)

where the aerosol volume phase function is

$$P^i_P(\psi, \lambda, y) = \frac{1}{2k^2\beta^i_P(\lambda, y)} \int_{r_1}^{r_2} \eta(r, y) \left\{ i_1(\psi, \tilde{m}, x) + i_2(\psi, \tilde{m}, x) \right\} dr$$

(12)

and $i_1$ and $i_2$ are the Mie intensity functions and $k = 2\pi/\lambda$.

The sky radiance due to the molecules and aerosols in the volume element $dV$ is then given by

$$dB(\phi, \phi_s, \omega, \lambda) = H_0(\lambda) \sec \phi \left\{ F^i_M(\psi, \lambda, y) + F^i_P(\psi, \lambda, y) \right\} e^{-\tau_1 + \tau_2} dy$$

(13)

Integrating over all such elemental volumes, the total single scattered sky radiance is

$$B(\phi, \phi_s, \omega, \lambda) = G \int_0^\infty \left\{ F^i_M(\psi, \lambda, y) + F^i_P(\psi, \lambda, y) \right\} e^{-\tau(\lambda, y)D} dy$$

(14)

where

$$G = H_0(\lambda) \sec \phi e^{-\tau(\lambda, 0) \sec \phi}$$

(15)

and

$$D = \sec \phi_s - \sec \phi$$

(16)

Before discussing the inversion problem, the photographic solar aureole measurement (PSAM) technique will be described.
A photographic technique of making measurements of the solar aureole radiance is briefly described here. Figure 3 schematically illustrates the equipment. Photographs of the Sun's aureole are taken with a small format camera (35 mm or 70 mm) through a wavelength filter with the Sun occulted by a neutral density (ND) disc (ND = 4) held coaxially on a stem about 0.6 m to 1.3 m in front of the lens. The ND filter attenuates the radiance of the direct sunlight by a factor of $10^4$, and the optical densities of the surrounding aureole are shown in a typical photograph in figure 4a. The Sun's image not only enables one to calibrate the entire photograph relative to its radiance, but also enables accurate measurements to be made of the angular distances from the Sun. Photographs are taken through different wavelength filters. In addition, the direct solar irradiance measurements through the same filters are made with a photometer to determine $\tau(\lambda, 0)$.

**A. Solar Aureole Isophotes**

The optical density of the whole photograph is read with the help of a Joyce-Loebl Isodensitracer, which gives digital data output on a magnetic tape and, at the same time, provides an isodensity tracing, such as shown in figure 4b. Isodensities are lines or curves of equal optical density in a photograph. Isophotes are then generated from the taped data output, as shown in figure 4c, where an economy-wise reduction has been made in the number of data points.

**B. Almucantar Radiance**

The photogrammetry of the solar aureole is discussed in detail in reference 21. It is shown that the almucantar projects on the film as a conic (fig. 5). The shapes of the conics for different values of the solar zenith angle $\phi_s$ are illustrated in figure 6. Accordingly, the measured almucantar radiance as a function of the scattering angle is shown in figure 7. The peak at zero degrees corresponds to the Sun's direct light reduced in intensity by the ND disc. With equation (14), it is possible to obtain the information about the size-altitude distribution of aerosols from the multispectral measurements of the sky radiance, $B(\lambda)$, as a function of $\phi_s$, $\phi$, and $\omega$, and of total optical depth $\tau(\lambda, 0)$. 
Many techniques for obtaining the aerosol characteristics suggest themselves in the light of this analysis. One of the simplest and most elegant is the method based on the almucantar radiance measurements, as explained in the following section.

**ALMUCANTAR RADIANCE AS AN INDICATOR OF SIZE DISTRIBUTION**

In the almucantar, which is a conical scan of constant solar zenith angle $\phi_s$ with the local zenith as axis, the total radiance $B$ is equal to the sum of radiance due to air molecules ($B_M$) and particulates ($B_p$), so that

$$B_p(\psi, \lambda) = B - B_M$$

where $\psi$ is the scattering angle, and $\lambda$ is the wavelength of solar radiation. From equation (17), one can then experimentally obtain the volume scattering function $F_p(\psi, \lambda)$ as a function of $\psi$, the quantity $F_p$ being a functional of $n(r)$. It is the experimental $F_p(\psi, \lambda)$ vs. $\psi$ data which is inverted to obtain $n(r)$. The experimental data for the altitude-integrated VSF is then given by

$$F_p(\psi, \lambda) = \frac{B(\phi = \phi_s, \omega, \lambda)}{G} - P_M(\psi, \lambda) M(\lambda, 0)$$

**RETRIEVAL OF AEROSOL SIZE DISTRIBUTION FROM ALMUCANTAR MEASUREMENTS**

Retrieval aerosol size distribution from equation (17) can be accomplished by either the model-fitting approach or the numerical inversion scheme.

**A. Model Catalog Search Method**

In this approach, an analytic model for $n(r)$ (ref. 22) is chosen from a parametrized catalog of phase functions which is generated for different values of $\bar{m}$ (ref. 23). As an illustration, the average value of $\bar{m}$ is assumed to be $1.55 - i(0.0)$, and a typical model for $n(r)$ is chosen as
\[ n(r) = \frac{p_3}{2} \left[ \frac{(p_1/r)^{v-3}}{p_2 \left( (p_1/r)^{v-3} - 1 \right) + 1} \right]^{1/2} \]

where \( p_1, p_2, p_3, \) and \( v \) are adjustable constants. Typical size distribution curves for different values of the parameter \( v \) are shown in figure 8. Any other analytic model of \( n(r) \) could do just as well.

A set of phase function curves corresponding to values of \( v \) that range from 4.0 to 5.2 are shown in figure 9. By comparing the experimental phase function curve in figure 8 with one of the curves in the catalog, one obtains a reasonably good estimate of the \( n(r) \). If, however, one wants to go a step further and obtain the \( n(r) \) that gives the best fit to the experimental phase function curve, a nonlinear least squares computer code is used. This is described next.

**B. Numerical Inversion Scheme**

The experimental \( F_p(\psi, \lambda) \) vs. \( \psi \) data was inverted by the use of nonlinear least squares (NLLS) method. On the basis of the exact Mie theory, \( F_p(\psi, \lambda) \) is defined by

\[ F_p(\psi, \lambda) = \frac{1}{2k^2} \int_{r_1}^{r_2} (i_1 + i_2) n(r) dr \]

where \( i_1 \) and \( i_2 \) are Mie intensity coefficients (ref. 18) \( r_1 = 0.04 \) \( \mu \)m and \( r_2 = 0.3 \) \( \mu \)m. The values \( i_1 \) and \( i_2 \) are computed using J.V. Dave's computer code. A two-term model of \( n(r) \) was selected with adjustable parameters \( p_i \), \( i = 1, 2, 3, \) and 4. Each term is a Haze M model (ref. 23), so that

\[ n(r) = p_1 r \left\{ \exp(-p_2 \sqrt{r}) + p_3 \exp(-p_4 \sqrt{r}) \right\} \]

Initial estimates of the parameters are obtained with the help of a parametrized graphical catalog of phase function plots (ref. 24) corresponding to different size distribution models (ref. 22). The best fit values of \( p_i \) are obtained by the use of the NLLS computer program. The method used in this program is
based on Taylor's series expansion of the function in terms of its parameters about the current estimates of the parameters and iteration of the resulting linear approximation until the estimates converge in the least squares sense. Results of typical inversion of experimental data for \( \lambda \) of 600 nm, 500 nm, and 400 nm, based on SS, are shown in figure 10. The experimental measurements were obtained during a field experiment described in section II.

RETRIEVAL FROM SOLAR AUREOLE ISOPHOTES

An extensive parametric computation of the solar aureole isophotes as functions of \( n(r) \) and \( \rho_p(y) \), and to some extent of \( \tilde{m} \), has been carried out by using equations (14) and (19). Everything else being the same, circumsolar isophotes corresponding to three values of the parameter \( v \) of the size distribution \( n(r) \) (fig. 7) are shown in figures 11a, 11b, and 11c. It is easy to see from figures 7 and 11 that, by slightly increasing the number of smaller particles and decreasing the number of larger particles as \( v \) becomes larger, the shape of the isophotes undergoes a dramatic change. As the value of \( v \) increases upwards from a value of 4.0, the isophote pattern attains a shape increasingly similar to that of the experimentally obtained isophotes (fig. 4c), until for a value of \( v = 5.0 \) the computer-generated pattern (fig. 11c) best resembles the latter. Thus, figure 11a illustrates the fine sensitivity of the patterns of the circumsolar isophotes to the size distribution \( n(r) \).

Attempts are underway to optimize computer programs that will obtain the best fit to the isophote pattern or numerically invert the isophote data by utilizing, to their advantage, the fact that radiance along each isophote remains constant. In this regard, it is important to keep in mind that, even though isophotes depend on both \( n(r) \) and \( \rho_p(y) \), within the aureole region the isophote shape is more sensitively dependent on \( n(r) \) than on \( \rho_p(y) \). The sensitivity of the isophotes to \( \rho_p(y) \) increases for larger angular distances from the Sun.

CONCLUDING REMARKS

In contrast to the modeling approach, in which one starts with a model judiciously hypothesized from experience, one begins the numerical inversion
approach with an initial estimate of parameters. A typical iterative scheme is described by Malchow and Whitney in reference 25. Simulations of sky radiance isophote shape or the phase function are performed by using equation (14) or equation (20), respectively, along with the initial set of parameters. Comparisons are made with the appropriate measurements. If the discrepancy is greater than a minimum prescribed value, then a parameter updating algorithm is used to obtain a new estimate of parameters. However, when the convergence criterion is satisfied, the final estimates of parameter are assumed to be accurate. This is very similar to the NLLS scheme.

It should be clearly pointed out that, since the radiative transfer program is called several times during each iteration in such an interactive inversion scheme, use of the Mie theory computations of scattered radiance in an inversion scheme can often become prohibitively expensive. Thus, in developing a radiative transfer code for use as an inversion scheme, it is imperative that it be computationally as fast as possible in order to be of practical use in numerical retrievals of aerosol characteristics.
II. PARTICIPATION IN UNIVERSITY OF ARIZONA-AEROSOL AND RADIATION
EXPERIMENT (UA-ARE), MAY 1978

The principal investigator, in concert with R.R. Adams of NASA/Langley
Research Center, participated in a multi-university, multi-instrument coor-
dinated aerosol and radiation experiment (ref. 26), conducted by Dr. J. Reagan
at the University of Arizona in May 1977. The solar aureole photographic
equipment was taken to Tucson, Arizona, and over 500 photographs of the Sun's
aureole were taken from the rooftop of the Engineering Building. Aureole
photographs were also taken at various altitudes (2000 to 9000 ft; 609.6 to
2,753.2 m) as we drove up to Mt. Lemón on the Catalina Mountains west of Tucson.
In addition, radiometer readings of the direct solar radiation were taken through
the same spectral filters that were used for photography.

These experiments were performed at the same time the other radiation
measurements, such as Sun radiometers, diffuse radiation measurement instru-
ments, bistatic lidar, were being deployed, thereby providing not only con-
sistency checks but also intercalibration of the data. Aerosol and radiation
measurements were also taken from aboard the Pennsylvania State University's
instrumented plane.

The nonlinear least-squares scheme described earlier was used to invert
the solar aureole data for \( \lambda = 400, 500, \) and 600 nm. The retrieved results
are summarized as follows (ref. 19):

The size distribution model was the sum of two Haze M models [eq. (21)].
Computations were performed in order to understand the effect of the following
parameters, viz, the refractive index, integration limit \( r_1 \) and, ignoring
multiple scattering, the retrieved size distribution. The results are
described as follows:

Three refractive indices were used, namely, 1.45 - i(0.0), 1.50 - i(0.0),
and 1.50 - i(0.01). Essentially no difference was found in the retrieved
results obtained for the different refractive indices, although the param-
eters were slightly different. In addition, the decrease of \( r_1 \) from 0.04
\( \mu \text{m} \) to 0.01 \( \mu \text{m} \) made no difference in the final \( n(r) \) results. For scattering
angles greater than 12 degrees, it was found that multiple scattering
should be taken into account to improve the accuracy of results.
Assuming single scattering, size distribution results were obtained by inverting the three sets of experimental data (fig. 10), first individually (ref. 26), and then simultaneously. Comparison among the retrieved size distributions obtained individually shows that the SD's for $\lambda = 0.5 \, \mu m$ and $0.6 \, \mu m$ are closer to each other than to SD for $\lambda = 0.4$. The main differences occur in the region around 1.0 $\mu m$, with SD for $\lambda = 0.4 \, \mu m$ having fewer particles (fig. 10). The correct method of obtaining the SD is by inverting the three sets of data simultaneously. The results thus obtained agree well with those obtained individually and also with the ground truth data obtained by airborne Whitby (⊗) and Royco (+) counters, which have been plotted in figure 10. Even though the ground truth data is sparse and is for May 7, 1977, the day following May 6, 1977, the day on which solar aureole measurements were made, they were reasonably close to the retrieved results and gave us confidence in the latter.
III. MULTISPECTRAL SOLAR RADIOMETRY FOR DETERMINING AEROSOL SIZE DISTRIBUTIONS

Measurements of the total optical depth $\tau(\lambda)$ of the atmosphere were made by measuring the direct solar radiation at various wavelengths $\lambda$ by means of a solar radiometer. These measurements can be inverted—indiscriminately or in conjunction with solar aureole data—to retrieve aerosol size distribution. We used a Tektronix radiometer (half-cone angle ~ 4 degrees) in our experiments. Whenever such a radiometer is used to measure the direct beam from a light source (the Sun in our case), some diffuse sky radiation invariably enters the detector's field of view along with the direct beam. If $I_0(\lambda)$ is the unattenuated intensity and $I(\lambda)$ is the intensity after passing through a medium, then the optical depth $\tau(\lambda)$ is given by Bouguer's (or Beer-Lambert's) transmission law

$$I(\lambda) = I_0(\lambda) e^{-\tau(\lambda)}$$

which is valid only for the direct radiation.

The experimentally measured intensity $I(\lambda)$, however, contains not only the direct beam, but also some forward-scattered radiation. Therefore, the optical depth obtained by the application of Bouguer's law is strictly speaking not the true optical depth, but an apparent optical depth. Thus, a forward-scattering correction factor $R$ is often used to account for this scattered radiation entering the detector's field of view. In order to understand the behavior of such a correction factor as a function of the particle size distribution and the complex refractive index, we carried out a theoretical parametric study first for the monodisperse aerosols (ref. 27) and then for polydisperse aerosols (ref. 28). These two references are included as Appendixes 1 and 2 of this report. The aerosol medium was assumed to be vertically and horizontally homogeneous. In this study, only single scattering was considered; multiple scattering was treated as being negligibly small. (It should be pointed out here that the general case in which multiple scattering and vertically inhomogeneous atmosphere are present will be treated in reference 29 as part of another NASA contract.)

Some of the conclusions of this investigation are summarized here as follows. It is shown that the forward-scattering correction factor, $R$,
decreases as the peak of the size distribution decreases. In addition, as the polydispersity of the size distribution becomes greater, the relative proportion of larger particles increases, resulting in an increase in the forward scattering of radiation within narrower angles. Other conclusions are that the lens-pinhole type detector optics, in most practical cases, yields more accurate transmission/extinction results than a detector without a lens-pinhole system. The conditions for optimum designs of radiometer/transmissometer detector systems are discussed in Appendixes 1 and 2 for the cases of monodisperse and polydisperse aerosol media, respectively. As a typical example, if we define error factor $E = (1 - R)$, then the percentage error contours representing trade-offs between particle size parameter $x(= 2\pi r/\lambda)$ and the detector viewcone half-angle $\theta$ degrees are shown in figure 12 for $m = 1.50 - i(0.0)$. 
IV. AEROSOL PHYSICAL PROPERTIES OF THE STRATOSPHERE (APPS) EXPERIMENT
(THE DRAPER LABORATORY AAFE PROJECT)

The principal investigator, Adarsh Deepak, assisted the technical monitor in Phases I and II of the APPS experiment developed under the AAFE project by the C.S. Draper Laboratory. Phase I involved the development of the theoretical algorithms and computer codes for inverting the spectral measurements, and Phase II involved the design of the instrument.

The development of the APPS experiment was aimed at the determination of the vertical profiles of atmospheric constituents and aerosol physical properties about 10 km altitude in the stratosphere and lower mesosphere. The experimental plan was to make simultaneous multispectral measurements of sunlight scattered by the Earth's atmospheric limb by a photometer at various narrow spectral bands from aboard the Space Shuttle platform, and to develop computer codes to invert these measurements for retrieving vertical profiles of the concentrations of Rayleigh scatterers, ozone, and aerosol extinction, aerosol size distribution, and real and imaginary parts of the complex refractive index. (These results are described in reference 25 by Malchow and Whitney.)

The theory for solving the radiative transfer equation for a spherical aerosol atmosphere and for the inversion problem is very complicated. However, theoretical studies performed by the author helped in redirecting some of the theoretical efforts being performed at the Draper Laboratory to yield satisfactory results within a brief period. The computer code, namely, Sunlit Limb Inversion Code (SLIC), developed at Draper Lab, was adapted for use on the LaRC computers. The lengthy computer programs were first checked out and later modified to include plotting capabilities. Some of the bugs in the program were removed and the programs were made more sophisticated.

A typical plot showing the inversion of the simulated data for 13 km to 24 km for an eight-channel sunlit limb radiance profile to obtain aerosol extinction profile is shown in figure 13. In the figure, circles represent the true state; squares, the state based on initial guess values of the parameters; diamonds, the state after the first iteration; and triangles, the final state after 2 to 10 iterations at different altitudes. The convergence to the true state for a maximum of 10 iterations seems rather good. It should improve for a larger number of iterations.
V. STRATOSPHERIC AEROSOL PHOTOGRAPHIC EXPERIMENT (SAPE)

The theory and inversion methods involved in SAPE are essentially the same as those in the APPS experiment, except that the multispectral measurements of the sunlit limb radiance profiles are obtained with a photographic camera rather than with a photoelectric radiometer. Whereas the APPS experiment is based on the measurements through eight spectral channels located in the near UV, visible, and near IR, SAPE involves measurements of limb radiance and its polarization through two wavelength filters.

The overall objective of the SAPE is to demonstrate the capability of the method to determine the stratospheric aerosol characteristics (physical and spatial) from the measurement of the solar radiation scattered by the Earth's atmospheric limb when the Sun is in any known position using sophisticated inversion techniques. If the Sun is within the limb, then the direct attenuated solar radiation is measured in addition to the forward-scattered radiation, so that inversion of the two sets of data yields results about aerosol characteristics that complement each other. It is the absence of restrictions upon the Sun's location with respect to the observed portion of the sunlit limb that gives this technique a temporal and global coverage that is greater than that achieved by any other existing technique.

The experimental objectives of SAPE are to measure, photographically, the profiles of (a) limb-scattered radiance and polarization, and (b) limb-attenuated direct solar irradiance and Sun-shape through narrow spectral regions at 880 nm and 650 nm, when the Sun is at various spacecraft (s/c) zenith angles. The exposure times needed to take analyzable photographs of the sunlit limb through the two filters have been evaluated. The photogrammetry, sensitometry, densitometry, and the photometry needed to obtain accurate radiance data have been extensively investigated.

Computer programs for inversion schemes required for retrieving aerosol and gaseous characteristics have been optimized. The anticipated results of SAPE are the profiles of stratospheric aerosol extinction, number density, size distribution, complex refractive index, and atmospheric refractive index. In addition, it is anticipated that detection of high-altitude particulate layers at 50 and 95 km will be made possible. The one thing that we need now is to obtain one or more photographs of sunlit limb from aboard the Space Shuttle.
VI. THE UCLA TROPOSPHERIC POLARIMETER AND MULTISPECTRAL RADIOMETER PROJECT

(THE POLARIMETER AND THE MULTISPECTRAL RADIOMETER AS
REMOTE PROBES OF AEROSOLS)

This project involved the determination of the tropospheric aerosol characteristics from the measurements of the polarization of scattered solar radiation in the Earth's turbid atmosphere. The measurements of scattered intensity and degree of polarization were performed with a NASA-funded polarimeter at UCLA by Dr. J. Kuriyan (ref. 30). It has been shown by several researchers that polarization of scattered radiation is sensitive to the aerosol refractive index. The inversion of the polarization data to obtain information about the aerosol properties is quite a formidable problem. So far, only the catalog search technique has been used (by Dr. J. Kuriyan) to retrieve aerosol SD and refractive index information. Numerical inversion schemes for polarization data have been investigated by the author, but a computer program has not yet been completed. The author plans to continue to develop such an inversion code. In addition, work on the sensitivity analysis of a Space Shuttle-based photopolarimeter technique was initiated at UCLA. The study was aimed at finding the optimum way in which the polarization measurements of upwelling radiation should be performed from aboard the Shuttle for global monitoring of the atmospheric aerosols. The algorithm for making such measurements and inverting the polarization data was based on the catalog search method. However, there is a strong need to develop a numerical inversion scheme for retrieving aerosol characteristics from polarization measurements of both upwelling and downwelling radiation.
VII. INTERNATIONAL INTERACTIVE WORKSHOP ON INVERSION METHODS IN ATMOSPHERIC REMOTE SOUNDING, WILLIAMSBURG, VIRGINIA, DECEMBER 15-17, 1976

This workshop was conducted under contract/task authorization NAS1-14193-24 as a part of grant NSG 1252. It was organized and chaired by the principal investigator, Adarsh Deepak, in cooperation with LaRC Technical Monitor, Dr. M.P. McCormick, who was the Associate Chairperson.

The ODU-Langley Research Center Workshop on Inversion Methods in Atmospheric Remote Sounding was a resounding success. Professor J. Lenoble, Université de Lille, France, reported on the Workshop in Applied Optics (ref. 31). The workshop attendance, which was by invitation only, consisted of 73 participants from seven countries, representing universities, research laboratories, and U.S. Government agencies. The presentation of invited papers by some of the world's leading experts and the interactive discussions made the workshop a very stimulating and valuable experience for everyone.

The workshop provided an interdisciplinary forum to review and assess the state-of-the-art in inversion methods available for retrieving information from remotely sensed data. The emphasis of the invited papers and the followup discussions covered the assumptions, methodology, resolution, stability, accuracy, and the future efforts needed in the various inversion methods. In this regard, the direct radiative transfer methods and results relevant to the inversion problem were also briefly reviewed in a special session on radiative transfer, held jointly with the OSA Topical Meeting on Atmospheric Aerosols, which preceded the workshop. This allowed researchers in different areas of remote sounding to compare and optimize the utilization of these inversion procedures in their respective remote sounding techniques. The workshop provided an avenue for free exchange of ideas and information among researchers concerned with the various inversion methods.

Twenty-two invited speakers presented papers covering the mathematical theory of inversion methods and the application of these methods to the remote sounding of atmospheric temperature, relative humidity, and gaseous and aerosol constituents.

The invited papers and the recorded discussions from the radiative session and the inversion method sessions of the International Workshop were edited by
VIII. MODELING OF AEROSOL PROPERTIES

A parametric study to develop graphical catalogs of (a) various analytic methods for aerosol size distribution and cumulative size distribution and (b) aerosol optical properties was initiated under grant NSG 1252. Work on the first catalog has been completed (ref. 22) and will shortly appear as a NASA publication. It is intended to be of great help to aerosol researchers in representing their experimental data by commonly used analytic functions. The study further shows the equivalence of analytic functions in best fitting the experimental size distribution (SD) data. The work on the second catalog (ref. 23), viz, for the optical properties associated with each of the size distributions is not yet complete. It will be completed under another contract. The computation of the optical properties is based on Mie theory computer codes. The optical properties of interest include the phase function and the extinction coefficient.

Eight analytic models for \( n(r) \) were considered. Their properties are summarized in table 1. A typical parameterized graph of \( n(r) \) vs. \( r \) for model 2, viz,

\[
n(r) = \left( \frac{p_1}{p_2} \right)^{\frac{p_3-1}{p_4}} \left( \frac{r}{p_2} \right)^{p_3} \left( 1 + \left( \frac{r}{p_2} \right)^{p_4} \right)^{-1}
\]

is shown in figure 14. In table 1, \( n(r) \) represents the size distribution \( \text{cm}^{-3}, \text{um}^{-1} \); \( N(r) \), the cumulative size distribution, \( \text{cm}^{-3} \); \( r_m \), the mode radius; \( M_k \), the kth moment; \( p_i \), \( i = 1,2,\ldots,n \) the model parameters; and the equation numbers mentioned in the table refer to the equations in reference 22 (to be published in the near future as a contractor report).
CONCLUDING REMARKS

Following are some of the important conclusions that emerged from the investigations described earlier.

The solar aureole photographic method for determining atmospheric aerosol characteristics is a simple and practical method, whose validity has been well demonstrated. As a result of these investigations, a number of publications have been issued by the principal investigator in co-authorship with coworkers at the Institute of Atmospheric Optics and Remote Sensing and NASA/Langley Research Center. In addition, several other groups around the world have started using this technique for the measurement of aerosol characteristics.

Our results on the forward-scattering corrections to optimum extinction measurements have been successfully checked out by Professor Allan Carswell, York University, Toronto, Canada, and his graduate student (as mentioned to the author in a private communication). These results were also helpful in interpreting NASA/LaRC's Tektronix radiometer data for solar radiation, obtained in conjunction with the solar aureole experiment.

The SLIC code developed for inverting horizon radiance profiles to retrieve stratospheric aerosol and gaseous characteristics is an extremely fast code, which takes account of multiple scattering in a spherical atmosphere. It needs further work done on it in order to make it more versatile so that extinction data as well as polarization data can also be inverted.

Expertise has been gained in the various aspects of solar aureole photography, as well as in the theory and inversion schemes investigated in connection with SAPE, to the point where now some photographs of the sunlit limb are needed from aboard the Space Shuttle.

Polarimetry of the upwelling and downwelling radiation provides a powerful technique for determining the aerosol characteristics. In order to make it a practical remote sensing method, a fast computer code for numerically inverting polarization data needs to be developed. The principal investigator intends to continue to develop such a code, which is not too far from completion.

There are three main components to remote sensing, namely, (a) the mathematics; (b) the physical concepts; and (c) the measurement techniques.
Much of the mathematics of the inversion problem were worked out in the 1960's. In order to make further improvements in accuracy of remote soundings, one needs to look more deeply into the physics and measurements aspects. The 1976 Williamsburg Workshop on Inversion Methods summarized many of the inversion techniques. It is suggested that a symposium or workshop dealing with the physics and measurement aspects of remote sensing be held in the near future.

The parametrized catalog of aerosol size distributions has proved quite valuable in representing the experimental size distribution data by analytic models. The completion of the corresponding parameterized catalog of the optical properties will be very useful in various aspects of aerosol research. The principal investigator intends to continue to work toward its completion.
APPENDIX 1

FORWARDSCATTERING CORRECTIONS FOR OPTICAL EXTINCTION MEASUREMENTS IN AEROSOL MEDIA. 1: MONODISPERSIONS
Forwardscattering corrections for optical extinction measurements in aerosol media. 1: Monodispersions

Adarsh Deepak and Michael A. Sox

This paper, Part 1 of two papers, presents a parametric study of the forwardscattering corrections for experimentally measured optical extinction coefficients in homogeneous aerosol media, since some forwardscattered light invariably enters along with the direct beam, into the finite aperture of the detector. Part 1 treats the case of monodispersions; Part 2, that of polydispersions. Forwardscattering is considered a single-scattering phenomenon, and the corrections are computed by two methods: one, using the exact Mie theory, and the other, the approximate Rayleigh diffraction formula. A parametric study of the dependence of the corrections on the particle size parameter, real and imaginary parts of the complex refractive index, and the half-angle of the detector's view cone has been carried out. The parameter ranges in which the results obtained by the approximate formulism agree well with those obtained by the Mie theory are also investigated. The agreement is especially good for small view cone angles and large particles and improves even more for slightly absorbing aerosol particles. Also discussed is the dependence of these corrections upon the experimental design of the transmission measurement systems.

I. Introduction

The forwardscattering correction refers to the correction that must be made to the optical extinction measurements carried out in a scattering medium, due to the fact that along with the direct radiation some forwardscattered radiation invariably enters into the finite aperture of the detector. In fact, no known light measuring device exists that can measure the intensity of the direct beam of radiation, with the complete exclusion of the light scattered in a forward direction. In making extinction measurements, one may optimally design the experimental apparatus so that the effects due to the forwardscattered radiation (and stray light) are minimized, but still in order to get the true extinction measurement, one must calculate and, if significant, correct for the forwardscattering contribution.

The term forwardscattering is considered a single, not multiple, scattering phenomenon in which scattered radiation reaches the detector after being scattered only once by scatterers situated within the path of the direct radiation. The case of multiple scattering, which causes additional corrections to the optical extinction measurements, will be discussed in a subsequent publication.

Two methods—one exact and one approximate—for deriving the forwardscattering corrections to the transmission law for both absorbing and nonabsorbing monodisperse aerosols are discussed in this paper. These methods have been discussed by Gumprecht and Sliepecevich for nonabsorbing monodisperse aerosols in connection with the problem of arriving at the optimum experimental design characteristics for an extinction experiment. Excellent discussions of the forwardscattering corrections are also given in Refs. 4-6. Here, however, the exact Mie theory is given in a closed form solution, which increases not only the accuracy of the results but also the efficiency of the computations. The approximate method is based on the Rayleigh diffraction theory for large particles. Approximate expressions for the correction factor at the small Rayleigh particle limit are also given. The discussions in this paper are confined to homogeneous spherical aerosols, having uniform size distribution, concentration, and composition, such as may occur along horizontal paths in field or laboratory situations.

II. Transmission Law (Bouguer's Law)

An electromagnetic plane wave of wavelength λ and intensity \( I_0(\lambda) \) (W/cm²-μm), after traversing a distance \( L \) through a homogeneous aerosol medium (Fig. 1), has intensity \( I(\lambda) \), given by Bouguer's Law
where $\tau(\lambda)$, the optical thickness, is defined as

$$\tau(\lambda) = \beta_{\text{ext}}(\lambda) L,$$

and the total volume extinction coefficient $\beta_{\text{ext}}(\lambda)$ is

$$\beta_{\text{ext}}(\lambda) = \beta_{\text{ext}}(\lambda) + \beta_{\text{abs}}(\lambda) \ [\text{cm}^{-1}],$$

the subscripts ext, scat, and abs denoting extinction, scattering, and absorption. For spherical monodispersions of number density $N_p(r) \ [\text{cm}^{-3}]$ and complex refractive index $m = m_r - i m_i$,

$$\beta_j = 4\pi Q_j(x,m)N_p(r), \quad j = \text{ext, scat, and abs},$$

where $x = 2\pi\lambda/\Lambda$ is the size parameter, and $Q_j$, the efficiency factors. If the molecular contribution to $\beta_{\text{ext}}$ is negligible, from Eqs. (3) and (4) one obtains

$$Q_{\text{ext}} = Q_{\text{ext}} + Q_{\text{abs}}.$$  \hspace{1cm} (5)

where

$$Q_{\text{ext}} = \frac{1}{2} \int_0^\infty (i_1 + i_2) \sin \theta d\theta,$$  \hspace{1cm} (6)

and $i_1$ and $i_2$ are functions of $\theta$, $x$, and $m$, called the Mie intensity functions.\footnote{Note that for the small values of $\theta$ considered in this work ($\theta \leq 10^\circ$), $i_1 \approx i_2$, and we may thus neglect the effects of polarization. Then a correction factor due to forwardscattering can be defined as}

$$R = Q_{\text{ext}} / Q_{\text{ext}} = 1 - E,$$  \hspace{1cm} (9)

where $E$, the error factor, is

$$E = \frac{1}{2\pi^2 Q_{\text{ext}}} \int_0^\infty (i_1 + i_2) \sin \theta d\theta.$$  \hspace{1cm} (10)

### B. Approximate Rayleigh Diffraction Formulation

Gumprecht and Sleipcevich\footnote{Gumprecht and Sleipcevich \cite{Gumprecht1978} have shown for small values of $\theta$ and large values of $x$ that as a good approximation to the exact Mie theory the value of $R$ can be computed from Rayleigh's diffraction theory formulation for large particles. Rayleigh has shown by diffraction theory that the fraction of the diffracted light falling outside a cone of half-angle $\theta$ in the forward direction is given by} have shown that for small values of $\theta$ and large values of $x$, the forward scattering correction and error factors are defined as

$$R = \frac{1}{2} [1 - J_0^2(\theta) - J_1^2(\theta)],$$  \hspace{1cm} (12)

$$E = \frac{1}{2} [1 - J_0^2(\theta) - J_1^2(\theta)],$$  \hspace{1cm} (13)

where $J_0$ and $J_1$ are Bessel functions of the first kind and of orders zero and one, respectively, $\theta$ is in radians and is small enough so that $\theta \approx \sin \theta$. We assume $Q = 2.0$ for large particles.

The Rayleigh diffraction approximation has been investigated by a number of authors, including Hodgkinson and Greenleaves,\footnote{Hodgkinson and Greenleaves also examined the error due to forwardscattered light using a formula similar to Eq. (13). In addition, they examined the geometrical optics effects of external reflection and transmission plus refraction, but their results indicate that these contributions are negligible for $\theta \leq 10^\circ$.} who also examined the error due to forwardscattered light using a formula similar to Eq. (13). In addition, they examined the geometrical optics effects of external reflection and transmission plus refraction, but their results indicate that these contributions are negligible for $\theta \leq 10^\circ$.

### IV. Corrected Transmission Law

By inserting the correction factor $R$ into the transmission equation, one can account for both the direct and forwardscattered radiation. Thus,

$$I(\lambda) = I_0(\lambda) \exp[-\tau(\lambda)],$$  \hspace{1cm} (14)

where

$$\tau(\lambda) = \pi L \int_0^L d\lambda' Q_{\text{ext}} R(x,\theta) N_p(r).$$  \hspace{1cm} (15)

Since the correction factor depends also on the geometry
of the transmission experiment, examples of two experimental designs are discussed in the next section.

V. Experimental Design Considerations

An extinction experiment essentially involves a source of radiation \( S \), a transmitter lens to transmit a collimated beam of known intensity \( I_0(\lambda) \) and radius \( R_3 \) through the aerosol medium of interest, and a detector system to measure the intensity of the direct beam after it has traversed a thickness \( L \) inside the medium. The correction factor \( R \) depends on the design of the measurement system, and, therefore, the design must be given careful considerations. In light of the discussions in the previous sections, two simple experimental systems are compared for their performance. Both use a collimated beam of radiation; they only differ in the design of their detector systems. One uses an open photodetector system [Fig. 1(a)] and the other, a lens–pinhole system [Fig. 1(b)]. The former will be referred to as simply the open detector system.

A. Open Detector System

The experimental system, shown in Fig. 1(a), consists of a radiation source, \( S \), situated at the focal length of a transmitter lens \( L_i \), which transmits a parallel beam through an aerosol medium of thickness \( L \), which is measured by an open photodetector of radius \( R_2 \). The detector subtends different angles \( \theta \) for scatterers located at different locations \( l \) inside the beam. Thus, the results differ for particles on and off the beam axis. These two cases are discussed as follows.

1. On-Axis Particles

For particles on the beam axis, angle \( \theta \) is given by

\[
\tan \theta = R_2/(L - l).
\]

(16)

In this case, since \( \theta = \theta(l) \), Eq. (15) can be written as

\[
\tau = L \pi r L_{\rho}(r) \rho_{\text{sm}}(x, \rho) R(x, L),
\]

(17)

where the path averaged correction factor \( \tilde{R} \) for a homogeneous aerosol medium is given by

\[
R(x, L) = \frac{1}{L} \int_0^L R(x, \theta(l)) dl,
\]

(18)

\[
\theta_0 = \tan^{-1}(R_2/L).
\]

In the exact formulation, the integral in Eq. (18) is solved by numerical quadratures. However, if one wishes to get some idea of how \( \tilde{R} \) behaves as a function of \( x \) and \( L \), it might be worthwhile to obtain an analytical solution of Eq. (18) under certain reasonable approximations. If one makes the approximations that \( \tan \theta = \theta \) and \( L \gg R_3 \) in Eq. (18), Eq. (18) reduces to

\[
R(x, L) = \frac{R_2}{L} \int_0^\infty R(x, \theta) e^{-\theta} d\theta.
\]

(19)

Using Eq. (12) and letting \( y = R_2x/L \), Eq. (19) yields

\[
\tilde{R} = \tilde{R}(y) = \frac{1}{2} - \frac{4y}{3} + \frac{1}{2} J_0(y) + \frac{1}{6} J_2(y)
\]

\[
+ \frac{2}{3} y^2 [J_0(y) + J_1(y)] - \frac{2}{3} y J_2(y) J_1(y).
\]

(20)

When \( y \to 0 \), \( \tilde{R} \) reduces to

\[
\tilde{R}_0 = 1 - 4y/3 + y^2/8 + O(y^4),
\]

and when \( y \to \infty \), \( \tilde{R} \) reduces to

\[
\tilde{R}_\infty \sim 1 + [2/(3\pi)] \cos 2y.
\]

In some experimental situations, there may be a gap between the scattering medium and the detector. In this case, the upper limit of integration in Eq. (18) will be less than \( \pi/2 \). Clearly, the integral can again be done numerically, but we may also use Eq. (20) if we subtract off the contribution to the integral due to the empty gap. For example, consider the scattering medium of length \( L \), and assume that the detector is located at a distance \( l_2 \) beyond the edge of the medium. Then it may easily be seen that

\[
\tilde{R}(L) = \frac{L + l_2}{L} \tilde{R}(L + l_2) - \frac{l_2}{L} \tilde{R}(l_2).
\]

(21)

2. Off-Axis Particles

For particles off the beam axis, the situation is clearly more complicated, as the cone subtended by the detector is no longer coaxial with the incident radiation. Thus, one can no longer assume the \( \phi \) integration implicit in Eq. (9), \( \phi \) being the azimuth. This additional complexity is beyond the scope of this work.

However, it is not too difficult to get some idea of the errors involved in ignoring this complication. Consider a particle at a location \( l \) along the beam and a radial distance \( \delta \) normal to the beam axis, as illustrated in the projection shown in Fig. 1(c). Here \( \delta < R_2 \). Then one may determine both the average scattering angle \( \bar{\theta} \) and the average projected distance \( \bar{d} \), where

\[
\tan \bar{\theta} = d/(L - l),
\]

\[
R_2^2 = \delta^2 + d^2 + 2\delta d \cos \phi,
\]

\[
\bar{\bar{\theta}} = \frac{1}{\pi} \int_0^\pi \left( \int_0^\pi \theta(\phi) d\phi \right) d\phi,
\]

\[
\bar{d} = \frac{1}{\pi} \int_0^\pi d(\phi) d\phi.
\]

Although \( \bar{\theta} \) cannot be determined analytically, \( \bar{d} \) can be expressed in terms of the complete elliptic integral of the second kind:

\[
\bar{d} = \frac{R_2}{\pi} \int_0^\pi \left( (1 - \rho \sin^2 \phi)^{1/2} - \rho \cos \phi \right) d\phi
\]

\[
= \frac{2R_2^2 E(\rho)}{\pi}
\]

(22)

where \( \rho = \delta/R_2 \). \( \bar{\theta} \) was evaluated numerically for a series of values of \( \rho \) and \( (L - l)/R_2 \), and it was found that, to a very good approximation in most cases, \( \tan \bar{\theta} = \bar{d}/(L - l) \). This approximation was found to be invalid only for the cases when \( \delta \) was large, and \( (L - l)/R_2 \) was small.
Thus, under most circumstances, the following equation is valid, namely:

$$\tan \theta = \frac{|R(x, \theta)|}{|\sigma(x, \theta)|}.$$  \hspace{1cm} (23)

When $\rho = 0$, $E = \pi/2$, Eq. (23) reduces to Eq. (16). The tables of $E$ may be used to infer typical magnitudes of errors in $\theta$, if Eq. (16) is used. Thus, when $\rho = 0.5$, $d = 0.934 R_2$, then the error in $\tan \theta$ is $\sim 6.6\%$ high; when $\rho = 1/\sqrt{2}$, $d = 0.860 R_2$, the error is $\sim 14\%$ high; and for $\rho$ as high as 0.9, $d = 0.745 R_2$, so that the error is about 25% high.

If particles are uniformly distributed out to some maximum value $\rho_{\text{max}}$ of $\rho$, the average value of $d$ for these particles is

$$D = \int \rho_{\text{max}} (A(\rho)) d\rho / \int \rho_{\text{max}} (A(\rho)) d\rho$$

$$= 4R \frac{E(\rho_{\text{max}}) + K(\rho_{\text{max}}) + E(\rho_{\text{max}})}{3 \sqrt{3}}$$  \hspace{1cm} (24)

where $K$ is the complete elliptic integral of the first kind.

One may now determine the average values of errors in $\theta$ and hence $\theta$, involved in ignoring off-axis particles. Thus when $\rho_{\text{max}} = 0.5$, $d = 0.968 R_2$, resulting in a 3.2% error; when $\rho_{\text{max}} = 1/\sqrt{2}$, $d = 0.914 R_2$, an 8.8% error; even when $\rho_{\text{max}} = 1$, then $d = 0.849 R_2$, resulting in an error of only 15.1%.

B. Lens–Pinhole System

The experimental system illustrated in Fig. 1(b) consists of a radiation source, $S$, situated at the focal point of a lens $L_1$, which produces a collimated beam of light. At the end of the beam path is a second lens $L_2$ of focal length $f$, which focuses the light through an aperture of radius $a$ in the focal plane and onto a detector. For such a system, it is easy to show that the half-angle $\theta$ is constant and is given by

$$\tan \theta = \frac{a}{d}.$$  \hspace{1cm} (25a)

provided the following condition holds, namely, that

$$R_2 \geq R_b + aL/f,$$  \hspace{1cm} (25b)

where $R_2$ is the radius of lens $L_2$. Since $\theta$ is constant for all particles anywhere inside the beam, for this system, referred to as the lens–pinhole system, Eq. (17) can be written as

$$r' = L \pi^2 N_p Q_{\text{ext}}(x, \theta) R(x, \theta),$$  \hspace{1cm} (26a)

since

$$R = R(x, \theta).$$  \hspace{1cm} (26b)

as given by Eqs. (9) or (12).

For particles whose distance from the beam axis $d$ and distance from the detector $d$ are such that $d + a/L > R_2$, the collecting lens $L_2$ may be considered a flat detector, and the results obtained earlier employed. Thus for circumstances in which Eq. (25b) does not hold, it is necessary to employ a combination of results.

VI. Computational Considerations

A parametric study of the correction factor $R$ and the corresponding percentage error $E$ was carried out as functions of different combinations of $x$, $\theta$, $m^*$, and $m^*$, whose values occur within the ranges $0.1 \leq x \leq 100$, $0^\circ \leq \theta \leq 10^\circ$, $1.33 \leq m^* \leq 1.65$, and $0.0 \leq m^* \leq 0.1$. The somewhat high values of $\theta$ and $m^*$, such as $\theta = 10^\circ$ and $m^* = 0.1$, were intentionally selected in order to understand the behavior of $R$ and $E$ as $\theta$ and $m^*$ approach these large values to gauge how valid are the approximations made in the theory in that region.

In the case of computations for $R$ and $E$ based on Mie theory, instead of employing a numerical quadrature scheme for evaluating the integral in Eq. (10), namely,

$$I(x, m, \theta) = \int_0^\theta \frac{(1 + i\sin \phi)}{(1 + i\sin \phi)^{n+1}} = \int_0^\phi (1 + i\sin \phi) d\phi,$$  \hspace{1cm} (27)

where $\mu = \cos \theta$ and $\mu' = \cos \theta'$, use was made of the following explicit closed form solution given by Wiscombe and Chylek [Eq. (4) in Ref. 11]

$$I(x, m, \theta) = \frac{1}{(1 - \mu^2)} [\text{Re}[R(x)] + \text{Im}[R(x)] - 2 \text{Re}[R(x)]R(x)]$$

$$+ \sum_{k=1}^{k-1} \left[ (2k + 1) \left[ a_k \right]^2 + b_k \right] H_k(\mu)$$

$$+ \sum_{k=2}^{k-1} \sum_{l=1}^{k-1} \left[ (l + 1 - k) b_l \right] H_k(\mu)$$

$$\times \left[ P_l(\mu) R_l(\mu) - P_l(\mu) R_l(\mu) \right] (1 - \mu^2),$$  \hspace{1cm} (28)

where

$$H_k(\mu) = \left( \begin{array}{c} 2k + 1 \left[ (2k + 1) a_k \right] - 2 \left[ (2k + 1) b_k \right] \end{array} \right)$$

$$R_l(\mu) = \left( \begin{array}{c} 2k + 1 \left[ (2k + 1) a_k \right] \end{array} \right)$$

and where $P_k$ is the Legendre polynomial, $R_l$ represents the complex conjugate of $R_l$, and $a_k$, $b_k$, and $\pi_k$ have their usual Mie theory meanings.

Then

$$E = I(x, m, \theta)/x^2 Q_{\text{ext}}.$$  \hspace{1cm} (30)

Computations for $R$ and $E$ by the approximate method were made by using Eqs. (12) and (13). Computations for the path-averaged correction factor $R$ were made for various values of $y$ by using Eqs. (12) and (20) for the lens–pinhole and open detector systems, respectively.

In Eq. (18), $\theta$ is a function of the two variables $x$ and $\theta_0$, which makes their plotting too complicated. In Eq. (20), however, $\theta$ is a function of $y$ only, which may be far more easily visualized. Equation (20) is, of course, only an approximation to Eq. (18), and it is important to determine the errors involved in using this expression. For this reason, Eq. (18) was evaluated numerically for a series of values of $x$ and $\theta_0$, and the results have been compared with those of Eq. (20).

The largest discrepancies occurred for large $\theta_0$ and for $x = 1.0$ to 2.0. For smaller values of $x$, both results were quite close to 1.0 for $\theta_0 \leq 20^\circ$, although the errors in $(1 - R)$ were fairly large. Note that the use of the Rayleigh expression in the evaluation of Eq. (18) probably...
introduces as much error as the approximation \( \tan \theta \approx \theta \) for \( x \) values in this range.

For \( \theta_0 = 20^\circ \), the largest discrepancy was 10.5% when \( x = 2 \). For \( \theta_0 = 10^\circ \), the largest discrepancy was 6.4%, while for \( \theta_0 = 5^\circ \), the largest discrepancy was only 2.2%. For smaller \( \theta_0 \) values, the errors decreased proportionally.

During this study, two other calculations were made. First, \( x \sin \theta \) was used instead of \( x \theta \) as the argument for the Bessel functions in the Rayleigh expression for \( R \). This produced virtually no effect on the values of Eq. (18). Second, the integral in Eq. (19) was terminated at \( \pi/2 \), instead of \( \infty \). This was found to produce considerable errors in almost all cases studied.

VII. Discussions and Conclusions

As mentioned earlier the parametric study of \( R \) was carried out as a function of many different combinations of \( x, \theta, m', \) and \( m'' \), but for the sake of clarity the results of only a few judiciously selected combinations are presented in the following subsections.

A. Results for \( R \) and \( E \) as Functions of \( x \) and \( m \)

Figures 2, 3, and 4 graphically depict the behavior of \( R \) and \( E \) as functions of \( x \) in the 0.1–100 range, for (1) three typical values of the view cone half-angle \( \theta \), namely, \( 1^\circ, 4^\circ, \) and \( 10^\circ \); (2) three typical values of \( m' \) namely, \( 1.33, 1.50, \) and 1.65; and (3) two complex values of \( m'' \), namely, 1.50–10.00 and 1.50–10.01. In reference to the plots \( E \) denotes percentage error obtained by multiplying \( E \) by 100; elsewhere, it denotes simply the error factor \( E \).

For smaller \( x \), Fig. 2 shows that for the three values of \( \theta \) and the five values of the refractive index, the value of \( R \) remains close to unity so that many details of the plots cannot be discerned. On the other hand, for the same small values of \( x \) the semilog plots of \( E \) vs \( x \) (Figs. 3 and 4) depict the behavior of \( E \) in much greater detail. The small particle limit behavior of \( E \) can be easily expressed by approximate analytic expressions as follows. As \( x \) values approach the small Rayleigh particle limit, one can use the lowest order approximations for \( l_1, l_2, \) and \( Q_{ext} \) given in Sect. 3.9 of Ref. 12 to derive the following approximate expressions for \( E \) for both the nonabsorbing and the absorbing particles, namely,

\[
E(m'' = 0) = \frac{1}{2} - \frac{3}{2} \cos \theta - \frac{3}{2} \cos^2 \theta \approx 3\theta^2/2. \tag{31a}
\]

\[
E(m'' \neq 0) = E(m'' = 0) \left[ \frac{x^3 (m'^2 - 1)^2}{m''} \right] \tag{31b}
\]

\[
E = \frac{\theta x^2 (m'^2 - 1)^2}{m''} \text{ for } m'' \ll 1. \tag{31b}
\]

Table I presents some sample approximate results, calculated with Eq. (31a) and (31b), along with the exact results, for \( \theta = 1^\circ \) and \( 10^\circ \), \( x = 0.1, 0.5, \) and 1.0, and \( m = 1.5-i(0.0), 1.5-i(0.02), \) and \( 1.5-i(0.1) \). From these results one can see that the approximate formulas are good to within a few percent of the exact results for \( x = 0.1 \) and are still reasonably good for \( x = 0.5 \). However, for \( x \leq 1.0 \), the behavior of \( E \) differs considerably from that predicted by Eq. (31), so much so that the \( m'' \) dependence virtually disappears for \( x \geq 3.0 \). It should be noted, however, that the \( \theta^2 \) behavior, predicted by Eq. (32) for both absorbing and nonabsorbing particles, is quite valid for \( \theta \leq 10^\circ \) and \( x \leq 5.0 \).
For large values of \( x(x ≥ 10) \) the effects of the various diffraction maxima and minima, or corona, can be clearly seen, particularly in Fig. 2. These effects are more important than the effects of refractive index for particles in this range, although it should be noted that the presence of some absorption tends to damp the oscillations. It should be pointed out that any attempt to repeat these graphs with a higher density of points in the \( 10 ≤ x ≤ 100 \) cycle merely results in a far more cluttered web. These oscillations disappear when the intensity is averaged over a suitable size distribution.

Figure 3 represents the behavior of \( E \) for nonabsorbing particles, and Fig. 4 represents that for particles with different values of the imaginary part of the refractive index. For nonabsorbing particles, the values of \( E \) rapidly reduce to constant threshold values \((~36^o/8)\) independent of \( x \) and \( m' \), as \( x \) reduces to less than about 1.0.

For absorbing particles, the behavior of \( E \) and \( R \) remain almost identical to that of nonabsorbing particles for this range of \( \theta \) and \( x > 1.5 \). However, for \( x ≤ 1.5 \), \( E \) decreases rapidly to values much lower than the threshold value for nonabsorbing particles. For very small absorbing particles the absorption dominates the scattering, \( E \) varying inversely as \( m'' \).

B. Results for \( R \) and \( E \) as Functions of \( \theta \)

Figures 5 and 6 illustrate the behavior of \( R \) and \( E \), respectively, as functions of \( \theta \) in the range \( 0.25^o \) to \( 10^o \) for \( m = 1.50 \) and for eight different values of \( x \) from 0.1 to 100. In Fig. 5, one sees that for \( x ≥ 5.0 \), the value of \( R \) falls off quite rapidly from unity, as \( \theta \) increases. The plots of \( E \) vs \( \theta \) in Fig. 6 depict the percentage errors due to forwardscattering for small values of \( \theta \) in greater detail. In Fig. 6, one sees that \( E \) increases as \( \theta \) increases for all values of \( x \), until it reaches a maximum value of about 0.5. The plots for \( x = 0.1 \) could easily be represented by Eqs. (31).

### Table 1. Comparison Between Small x Approximation and Exact Values for the Error Factor \( E(x,m,\theta) \)

<table>
<thead>
<tr>
<th>( m )</th>
<th>( x )</th>
<th>( \theta = 1^o )</th>
<th>( \theta = 10^o )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5-(0.00)</td>
<td>0.1</td>
<td>1.142(-4)</td>
<td>1.148(-4)</td>
</tr>
<tr>
<td>0.5</td>
<td>1.142(-4)</td>
<td>1.278(-4)</td>
<td>1.142(-2)</td>
</tr>
<tr>
<td>1.0</td>
<td>1.142(-4)</td>
<td>1.738(-4)</td>
<td>1.142(-2)</td>
</tr>
<tr>
<td>1.5-(0.02)</td>
<td>0.1</td>
<td>6.610(-7)</td>
<td>6.582(-7)</td>
</tr>
<tr>
<td>0.5</td>
<td>8.263(-5)</td>
<td>5.012(-5)</td>
<td>8.263(-3)</td>
</tr>
<tr>
<td>1.0</td>
<td>6.610(-4)</td>
<td>1.372(-4)</td>
<td>6.610(-2)</td>
</tr>
<tr>
<td>1.5-(0.1)</td>
<td>0.1</td>
<td>1.322(-7)</td>
<td>1.375(-7)</td>
</tr>
<tr>
<td>0.5</td>
<td>1.653(-5)</td>
<td>1.511(-5)</td>
<td>1.653(-3)</td>
</tr>
<tr>
<td>1.0</td>
<td>1.322(-4)</td>
<td>0.761(-4)</td>
<td>1.322(-2)</td>
</tr>
</tbody>
</table>

C. Error Contour Diagram

It is obvious from the results shown in Figs. 2-6 that one can plot a series of percentage error contours in the \( x - \theta \) plane to illustrate trade-offs that can be made between the values of \( x \) and \( \theta \). Three contours for percentage errors of 1%, 5%, and 10% are shown in Fig. 7 for \( m = 1.50-(0.00) \) and for the ranges \( 1 ≤ x ≤ 100 \) and \( 0^o ≤ \theta ≤ 10^o \). The 1% contour shows an inflection point in its curvature at about \( \theta ≈ 5^o \).
D. Comparison Between Mie Theory and Diffraction Formulations

The approximate expressions for $R$ and $E$ in Eq. (13) have no $m$-dependence. Figures 8–12 show plots of $R$ vs $\theta$, as computed using both Eqs. (11a) and (13a), for a series of values of $x$ and a series of refractive indices. Symbols without a cross represent exact Mie results [Eq. (11a)], and symbols with a cross represent Rayleigh diffraction results [Eq. (13a)].

Figures 8, 9, and 10 show that for nonabsorbing particles with three different refractive indices (1.33, 1.65, and 1.5), the Rayleigh diffraction results are in fairly good agreement (within about $\pm$ 5%) with the exact Mie results for a wide range of $x$ and $\theta$ values. However, for small values of $x$, while both expressions yield results close to unity for $R$, they may yield significantly different values of $E$. The agreement appears to be best for $\theta \leq 1.5^\circ$. 
For absorbing aerosol particles, Figs. 11 and 12 show that agreement improves rather dramatically for $\theta$ larger than 1.5° as $m^* \theta$ increases its value from 0.00 to 0.1. Even when the absorption is small, the agreement between results of the two methods significantly improves, especially for large values of $x$. This is an extremely useful result when absorbing aerosol particles are involved, as the use of the Rayleigh diffraction formula [Eq. (13a)] can yield accurate results, while at the same time considerably reducing computation costs. In addition, this approximation might be amenable to analytical integration in some cases, as shown earlier, which could be a valuable asset in the optimum designing of an extinction experiment.

E. Optimum Transmissometer Design

Equation (14) shows how one includes the forwards-scattering correction factor to obtain the measured (or apparent) optical depth $\gamma'$. Of the various optical systems that have been used in light-transmission experiments, two of which are described in earlier sections, the lens–pinhole optical system is preferred because the value of $\theta$ is constant and independent of the location of the illuminated particle inside the beam path, as long as Eq. (25b) is satisfied. In addition, the lens–pinhole system practically excludes all stray light from the detector.

In contrast to the lens–pinhole system, the open detector system [Fig. 1(a)] presents obvious difficulties, such as (1) $\theta$ is not constant but depends on the location of the scatterer in the beam path, so that computations for $R$ are somewhat cumbersome; and (2) the value of $R$ for particles near the outer edges of the beam differs from that for particles at the center of the beam at any given distance from the detector, due to the fact that the axis of the cone subtended by the detector with the former particle is not coincident with the direction of propagation of the incident light.

The values of the path averaged correction factor $\tilde{R}$ as given by Eqs. (20) and (26b) for the open and the lens–pinhole detector systems, respectively, are plotted in Fig. 6 as a function of $y (= x\theta)$, where $R_2/L$ and $\tan^{-1}a/f$ are the respective values of $\theta$ for the two systems. The values of $\tilde{R}$ for the open detector are smaller than those for the lens–pinhole system for the $0.01 < y < 100$ range. This difference in the two sets of values of $\tilde{R}$ tends to disappear for extreme values of $y$, viz., $y < 0.02$ and $y > 15.00$. For $y \rightarrow \infty$, $\tilde{R} \rightarrow 0.50$. These results are useful for designing transmission experiments. Maximum accuracy is achieved if one designs the experiment such that $\tilde{R}$ is nearly constant, which occurs at the extreme values of $y$. In these $y$ ranges, the lens–pinhole detector is more accurate than the open detector (see Fig. 6). However, in the intermediate $y$ region, the uncertainty in $\tilde{R}$ due to variations in $y$ for the lens–pinhole detector can be larger than that for the open detector due to the steeper gradient.

From the foregoing discussion, it is apparent that the transmission law as ordinarily expressed by Eq. (1) cannot be used except in the case of small particles, for which it is only a good approximation. The effect of not only the particle’s size and composition but also the geometry of the optical system on the apparent volume scattering and extinction coefficients must be considered.

The results discussed in this paper pertain to the case of a collimated beam of radiation traversing a homo-
Fig. 13. Plots of the path-averaged correction factor $\tilde{R}$ as a function of $y$, where $y = \pi a/l$ and $zR_3/L$ for the lens-pinhole detector and open-detector systems, respectively.

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References
APPENDIX 2
FORWARDCATTERING CORRECTIONS FOR OPTICAL EXTINCTION
MEASUREMENTS IN AEROSOL MEDIA: 2: POLYDISPERSIONS
Forwards scattering corrections for optical extinction measurements in aerosol media. 2: Polydispersions

Adarsh Deepak and Michael A. Box

This paper, second of two parts, presents a parametric study of the forwards scattering corrections for experimentally measured optical extinction coefficients in polydisperse particulate media, since some forward scattered light invariably enters, along with the direct beam, into the finite aperture of the detector. Forwards scattering corrections are computed by two methods: (1) using the exact Mie theory, and (2) the approximate Rayleigh diffraction formula for spherical particles. A parametric study of the dependence of the corrections on mode radii, real and imaginary parts of the complex refractive index, and half-angle of the detector’s view cone has been carried out for three different size distribution functions of the modified Gamma type. In addition, a study has been carried out to investigate the range of these parameters in which the approximate formulation is valid. The agreement is especially good for small-view cone angles and large particles, which improves significantly for slightly absorbing aerosol particles. Also discussed is the dependence of these corrections on the experimental design of the transmissometer systems.

I. Introduction

In Part 1 of two papers, results were presented of a parametric study of the forwards scattering correction factor \( R \) and the complementary error factor \( E \) for monodispersions. In this paper the results of a similar study carried out for spherical polydispersions of size distribution \( n(r) \) (cm\(^{-3}\) \( \mu \)m\(^{-1}\)), \( r \) being the radius in \( \mu \)m, will be presented. For the sake of clarity, only the results obtained with the use of simple unimodal size distributions of the modified Gamma type, such as Dermendjian models2 Haze M, Haze H, and Cloud C3 (referred to as Haze C in this paper), are presented here. Results for other real size distributions can easily be obtained in a similar manner. The behavior of both \( R \) and \( E \), averaged over each of the three size distributions, will be discussed here as functions of each of the following parameters: the mode radius \( r_m \); the polydispersity or the spread of the size distribution; and the real \( (m') \) and imaginary \( (m'') \) parts of the complex refractive index \( m = m' - im'' \). The computations have been carried out with both the exact Mie theory solutions in explicit closed form and the Rayleigh diffraction theory approximation, as explained in Part 1. The results of such a study are extremely useful in obtaining the optimum experimental design parameters for the measurement of extinction coefficients in particulate media.3

II. Transmission Law (Bouguer’s Law)

The transmission law for an electromagnetic plane wave passing through a homogeneous polydisperse aerosol medium (Fig. 1, Part 1) is given by Bouguer’s law, defined by Eq. (1) in Part 1, namely,

\[
I(\lambda) = I_0(\lambda) \exp\left[-\tau(\lambda)\right], \tag{1}
\]

where

\[
\tau(\lambda) = \beta_{\text{ext}}(\lambda)L, \tag{2}
\]

\[
\beta_{\text{ext}}(\lambda) = \beta_{\text{ext}}(\lambda) + \beta_{\text{abs}}(\lambda). \tag{3}
\]

As in Part 1, for the sake of clarity, the molecular contributions to \( \beta_{\text{ext}} \) will be ignored here. Then for polydisperse aerosols of size distribution \( n(r) \), cm\(^{-3}\) \( \mu \)m\(^{-1}\), the coefficients are defined by

\[
\beta_{j}(\lambda) = \int_{r_1}^{r_2} \pi r^2 Q_i(x,m)n(r)dr, \quad j = \text{ext, scat. and abs.} \tag{4}
\]

All the quantities in Eq. (4) are the same as defined in Part 1, except that the factor \( N_p(r) \) for monodisperse particles has been replaced here by the operator

\[
\int_{r_1}^{r_2} n(r)dr,
\]

where \( r_1 \) and \( r_2 \) are lower and upper limits of radii. Aerosol size distributions are discussed in a later section.
III. Forwardscattering Corrections

A. Exact Mie Theory Formulation

Because of the fact that forwardscattered light invariably enters the detector view cone, Bouguer's law, as defined in Eq. (1), cannot be used to obtain the true optical depth \( \tau \) in a transmission experiment. But instead, one obtains the apparent optical depth \( \tau' \) related to the apparent volume extinction \( (\beta_{\text{ext}}) \) and scattering \( (\beta_{\text{scat}}) \) coefficients, which are distinguished from the true quantities \( \beta_{\text{ext}} \) and \( \beta_{\text{scat}} \) respectively; here, \( \beta_{\text{scat}} \) is clearly not affected by the forwardscattering. Thus,

\[
\beta_{\text{ext}} = \beta_{\text{scat}} + \beta_{\text{abs}} \quad \text{(apparent). (5)}
\]

The Mie efficiency factor \( Q_{\text{scat}} \) is based on the total amount of light lost from the beam by scattering by a particle in all directions; and \( Q_{\text{ext}} \), on the amount of light lost by scattering in all directions except within a cone of half-angle \( \theta \) in the forward direction, so that their difference is given by

\[
Q_{\text{ext}} - Q_{\text{scat}} = Q_{\text{ext}} - Q_{\text{scat}} = \frac{I(x,m,\theta)}{2L} = \frac{I(x,m,\theta)}{2L}.
\]

where \( I(x,m,\theta) \) is defined in Eqs. (28) and (29) in Part 1. From Eqs. (3) and (5), one obtains

\[
\beta_{\text{ext}} = \beta_{\text{scat}} - \beta_{\text{ext}} = \beta_{\text{scat}} - \beta_{\text{ext}} \quad \text{and} \quad \beta_{\text{scat}} = \beta_{\text{scat}} - \beta_{\text{scat}}.
\]

B. Approximate Rayleigh Diffraction Formulation

From Eq. (12) in Part 1 and Eqs. (6) and (8), the Rayleigh diffraction approximation to the forwards-cattering correction for a polydispersion is given by

\[
\bar{R} = \frac{\pi}{\beta_{\text{ext}}} \int_0^{\infty} r^n(r)I(x,m,\theta)dr.
\]

where \( J_0 \) and \( J_1 \) are Bessel functions of the first kind and of orders zero and one, respectively.

IV. Corrected Transmission Law

By inserting the correction factor \( \bar{R} \) into the transmission equation, one can account for both the direct and forwards-scattered radiation. Thus

\[
I(\lambda) = I_0(\lambda) \exp(-\tau'(\lambda)), \quad (11)
\]

where

\[
\tau' = \int_0^L dl \beta_{\text{ext}} = \int_0^L dl \beta_{\text{scat}}[n(r,\theta)]
\]

From the discussion in Part 1, it is clear that for parallel beam transmission systems with an open detector, the half-cone angle \( \theta = \theta_{(l)} \), so that

\[
\tau' = \pi L \int_0^{\infty} drr^2Q(x,m)n(r)L(x,L).
\]

where

\[
R(x,L) = \frac{1}{L} \int_0^L dl R(x,\theta(l)).
\]

And for a lens–pinhole system \( \theta = \text{constant} \), so that \( R = R(x,\theta) \), independent of \( l \). For the discussion of the experimental design considerations of the two detector systems, see Part 1.

V. Aerosol Size Distributions

Several analytic representations of aerosol size distributions appear in the literature. In this paper, the behavior of \( \bar{R} \) and \( \bar{E} \) is investigated as a function of the mode radius \( r_m \), the spread of the size distribution and the complex refractive index \( m = m' - im'' \), the upper and lower limits of radii being \( 10^{-2} \) and \( 20 \mu m \). For the sake of simplicity, three of the Deirmendjian models, namely, Hazen M, and Cloud C3 (referred to here as Hazen C), were selected for representing different polydispersities of aerosol size distribution \( n(r) \). Since \( r_m \) is varied between the radii limits \( 10^{-1} \) and \( 10 \mu m \), the three models are used here in a more general way than was their original intent. It is in that sense that model Cloud C3 is referred to as Hazen C. The limits of integration over \( r \) are \( 10^{-2} \) and \( 20 \mu m \).

The expressions for the \( n(r) \) models and their corresponding mode radii \( r_m \) are given as follows:

(a) Hazen M: \( n(r) = r \exp(-br^4/2) \), \( r_m = 4/b \),

(b) Hazen H: \( n(r) = r^2 \exp(-br^2) \), \( r_m = 2/b \),

(c) Hazen C: \( n(r) = r^3 \exp(-br^3) \), \( r_m = (8/3)^{1/3}b^{-1} \).

Note that any normalization or scale factor in the size distribution will cancel when \( \bar{E} \) and \( \bar{R} \) are evaluated. Thus, although \( \bar{E} \) and \( \bar{R} \) do depend on the shape of the size distribution, they do not depend on the total number of particles.

The difference in the size of these three models is illustrated in Fig. 1, where each has \( r_m = 1.0 \mu m \) and \( n(r_m) = 1.0 \times 10^6 \mu m^{-1} \). For want of a better terminology, the spread of a size distribution will also be referred to as polydispersity of the size distribution in this paper. For example, Hazen H will be referred to as more polydispers than Hazen C, and in the same vein it will be stated that Hazen M has a higher polydispersity than Hazen H. It should be noted here that there exist other
terms in the theory of distribution functions to express the same quantity.

Perhaps the major disadvantage of the Deirmendjian models is that their falloff behavior for large radii is too sharp: many experimentally measured distributions show a power law behavior, at least in the optically active region. For this reason, we have also considered a power law distribution, given by

\[ n(r) = r^{-\nu}, \quad 10^{-3} \mu m \leq r \leq 15 \mu m \]

and allowed \( \nu \) to vary between 2 and 4. Note that one cannot talk about either mode radius or polydispersity for a power law haze, only slope \( \nu \).

**VI. Computational Considerations**

A parametric study of the correction factor \( R \) and the corresponding error factor \( E \) was carried out as functions of different combinations of \( r_m, \theta, m' \) and \( m'' \), whose values occur within the ranges \( 0.1 < r_m < 10 \mu m, \quad 0^\circ < \theta < 1^\circ, \quad 1.33 < m' < 1.65, \quad 0.0 < m'' < 0.1 \).

The computations of \( R \) (and \( E \)) in Eq. (8) [and Eq. (9)] are made by using the closed form relations given in Part 1 for \( I(x, m, \theta) \). The computations of \( R \) for the approximate method are made by using Eq. (10).

**VII. Discussion and Conclusions**

The parametric study of \( R \) was carried out as a function of many different combinations of \( r_m, \theta, m' \), and \( m'' \), but for the sake of clarity the results of only a few judiciously selected combinations are presented in the following subsections. As the Deirmendjian models are so different from the power law model, we shall treat each separately, starting with the Deirmendjian models.

**A. Results for \( R \) and \( E \) as Functions of \( r_m \) and \( m' \)**

Figures 2–7 illustrate the behavior of \( R \) and \( E \), respectively, as functions of \( r_m \) in the 0.1–10.0-\( \mu \)m range, for three different half-cone angles (1°, 4°, and 10°) and five different refractive indices [1.33, 1.65, 1.55–i(0.05), 1.55–i(0.1)], for each of the three models: (a)
Haze $M$, (b) Haze $H$, and (c) Haze $C$, respectively. The computations were made for $\lambda = 0.55 \, \mu m$.

For Haze $M$ (Fig. 2), the values of $\bar{R}$ rapidly decrease as $r_m$ increases from 0.1 $\mu m$ to about 1 $\mu m$ and then tend to level off to a nearly constant value. The percentage error (Fig. 5) for Haze $M$ shows that for small $r_m$ the error $E$ increases fairly rapidly and then quickly levels off to a constant value, which is different for each set of $\theta$ and $m$ values. The smaller the $\theta$ the higher is the value of $r_m$ beyond which the leveling of the values for $E$ takes place.

For Haze $H$ and Haze $C$, the plots of $\bar{R}$ make an inverted integral sign (2) within the $r_m$ range of 0.1–1.0 $\mu m$, their steepness increasing with the decrease in polydispersity of the size distribution. From Figs. 2–7, we see that for $\theta = 1^\circ$, there is virtually no $m$-dependence for any of the hazes. For this reason, very few symbols have been drawn on these lines. For $\theta = 4^\circ$, we see that a small $m$-dependence has started to appear, being most clearly visible in Figs. 5–7, due to the logarithmic scale for $E$. However, for $\theta = 10^\circ$, a clear $m$-dependence can be distinguished, especially in Figs. 2–4. We see, in general, that $\bar{R}$ is lowest for $m = 1.33$, and that there is little difference between the curves for $m = 1.55$ and 1.65. However, the presence of a small amount of absorption immediately raises $\bar{R}$, although the actual value of this absorption appears to have little effect.

Effect of polydispersity: Comparing the $\bar{R}$ (and $E$) vs $r_m$ plots for the three models, one can reach the following conclusions:

It can be seen from the plots presented here that, in general, for given values of $\theta$ and $r_m$, the higher the polydispersity the higher the error $E$. This is due to the predominant forwardscattering of the large particle component, implied by the increased polydispersity. We should note, however, that the saturation values of $\bar{E}$ and $\bar{R}$ (both $\approx 0.5$) are not affected by polydispersity, but that these values are reached sooner for a more polydisperse haze than for a less polydisperse haze. A saturation value of $\bar{E} \approx 0.5$ implies a $Q_{ext}$ value of 1.0. An explanation for this is as follows. For the same value of $\theta$, the amount of scattered radiation collected at the detector increases as the large particle component increases, which, in effect, reduces $Q$ from a value of about 2.0 (assumed for large particles) to a minimum value of 1.0. The saturation value for $\bar{E}$ tends to be about 50% for $\theta = 10^\circ$ and 45% for $\theta = 4^\circ$ for Haze $M$, a difference of about 5%. This difference, however, tends to decrease as aerosol size distributions become less polydisperse, implying a smaller number of large particles, so that the saturation value of $\bar{E}$ approaches 50%.

B. Results for $\bar{R}$ and $\bar{E}$ as a Function of $\theta$.

Figures 8–10 show the behavior of $\bar{E}$ as a function of $\theta$ for six values of mode radius (0.1, 0.15, 0.50, 1.0, 2.5, and 10.0 $\mu m$), and $m = 1.55$ for each of the three size distribution models, respectively. Again, the effects of polydispersity are quite apparent in the increasing spread of the $\bar{E}$ vs $\theta$ plots for lower $r_m$ values (below 1.0 $\mu m$) as the polydispersity decreases.

Figures 11–13 illustrate the behavior of $\bar{R}$ as a function of $\theta$ for five values of $r_m$ (0.1 $\mu m$, 0.4 $\mu m$, 1.0 $\mu m$, and 1.0 $\mu m$). Symbols without crosses represent the Mie theory results, and those with cross represent the approximate results. The discussion of their comparison will be presented in the next section. Only the Mie theory results will be discussed here. The
Fig. 8. Plots of percentage error (100%) vs half-angle $\theta$ for six mode radii ($m = 1.55$): Haze $M$.

Fig. 9. Plots of percentage error (100%) vs half-angle $\theta$ for six mode radii ($m = 1.55$): Haze $H$.

Fig. 10. Plots of percentage error (100%) vs half-angle $\theta$ for six mode radii ($m = 1.55$): Haze $C$.

Fig. 11. Plots of correction factor $\bar{R}$ vs $\theta$ for $m = 1.55$, $\lambda = 0.55 \mu m$, and five mode radii: Haze $M$. (Symbols without crosses represent Mie results; symbols with crosses, approximate results.)

Fig. 12. Plots of correction factor $\bar{R}$ vs $\theta$ for $m = 1.55$, $\lambda = 0.55 \mu m$, and five mode radii: Haze $H$. (Symbols without crosses represent Mie results; symbols with crosses, approximate results.)

Fig. 13. Plots of correction factor $\bar{R}$ vs $\theta$ for $m = 1.55$, $\lambda = 0.55 \mu m$, and five mode radii: Haze $C$. (Symbols without crosses represent Mie results; symbols with crosses, approximate results.)
choice of different \( r_m \) values for the cases shown in Figs. 8–13 was merely for the sake of the clarity of the graphical display. The plots in Figs. 11–13 also show that the correction factor \( R \) falls rapidly with increasing \( \theta \) for large \( r_m \) values and then levels off quickly to a value close to 0.5. For large particles, the plots are practically identical for all the three models. However, for small values of \( r_m \), there is a considerable spread in the values of \( R \).

C. Comparison Between the Mie Theory and Diffraction Formulation

Consider Figs. 11–16 representing the \( R \) vs \( \theta \) results for nonabsorbing \( (m = 1.55) \) and absorbing \( (m = 1.55 - i 0.05) \) aerosols, respectively. Symbols with crosses represent the results due to Rayleigh formulation and those without crosses the results due to Mie theory. Figures 11 and 14, 12 and 15, and 13 and 16 represent the models Haze \( M \), Haze \( H \), and Haze \( C \), respectively.

Figures 11–13 show that, for nonabsorbing aerosols, the agreement between the Mie and the Rayleigh approximation is not as good for small \( r_m \) as it is for larger \( r_m \) for all three models. This is to be expected since the Rayleigh diffraction formula Eq. (13) is only valid for large particles. On the other hand, the agreement for both large and small \( r_m \) appears to be better for Haze \( M \) (more polydisperse aerosol) than for Haze \( C \), indicating that for more polydisperse size dispersions the agreement between the Rayleigh formulation and the exact formulation tends to improve. Comparison of these results with those for monodispersions in Part 1 also substantiates the trend. The remarks for the case of nonabsorbing aerosols apply to the case of absorbing aerosols \( (m = 1.55 - i 0.05) \) as well, except for the additional conclusion that the agreements between the Rayleigh and Mie results for all models are considerably better for the absorbing aerosols than for the nonabsorbing (see Figs. 14–16). The latter conclusion is again in line with the one made for monodispersions in Part 1.

D. Error Contours Diagram in \( r_m - \theta \) plane

Figure 17 shows the error contours for 10% (dashed line) and 5% error (solid line) for \( m = 1.55 \) and models \( M, H \), and \( C \). The curves indicate an inflection point in Haze \( M \) (10%) curve and Haze \( H \) (5%) at about \( \theta = 1.5^\circ \) and \( \theta = 6^\circ \), respectively, in the range \( r_m \sim 0.1 \mu m \).

E. Experimental Design Considerations

Equation (11) shows how the forwards-scattering correction factor is included in the transmission law to obtain the measured (or apparent) optical depth \( \tau \). Comments about the two aforementioned experimental geometries, discussed in Paper 1, also apply to the case of polydisperse aerosols, in addition to the following comments regarding the effects of the polydispersity on the experimental design.

In Paper 1 (Fig. 13), results were plotted for \( R \) as a function of \( x\theta \) for the two aforementioned experimental geometries. For the polydisperse case, however, \( R(x\theta) \) has to be further averaged over the volume extinction coefficient to yield \( R \), the path-averaged correction.
Fig. 17. Error contours in the $r_m - \theta$ plane for $m = 1.55$, $\lambda = 0.55\, \mu m$, for models Haze M, Haze H, and Haze C.

Fig. 18. Plots of the path-averaged correction factor $\tilde{R}$ vs $y$, where $y = kr_m \theta / \sqrt{kr_m}$ for the lens-pinhole detector and open detector systems, respectively, for models Haze M, Haze H, and Haze C. $\lambda = 0.55\, \mu m$.

Fig. 19. Error contours in the $y - \theta$ plane for $m = 1.33$, $\lambda = 0.55\, \mu m$, for the power law size distribution.

Fig. 20. Error contours in the $y - \theta$ plane for $m = 1.55 - (0.05)$, $\lambda = 0.55\, \mu m$, for the power law size distribution.

Fig. 21. Plots of correction factor $\tilde{R}$ vs $\theta$ for $m = 1.55 - (0.05)$, $\lambda = 0.55\, \mu m$, and five values of the power $s$ for a power law size distribution. (Symbols without crosses represent Mie results; symbols with crosses, approximate results.)
In order to make accurate transmission measurements, it is important that the experimental transmission design be based on those values of \( R \) for which either \( R \to 1 \) or \( R \to 0.5 \). But to be able to do so, some reasonable prior knowledge of both the mode radius \( r_m \) and the polydispersity is required.

F. \( \lambda \)-Dependence of \( R \) and \( E \)

All the results shown in the various plots were computed for \( \lambda = 0.55 \mu \text{m} \). For any other value \( \lambda' \), these same results are valid for the three models, Haze \( M \), Haze \( H \), and Haze \( C \), provided \( r_m \) is replaced by \( r_m' = r_m/\lambda' \).

A more detailed parametric study of the behavior of the forwardscattering effects involved in different transmission measurement experiments designed to measure optical extinction and visibility in the atmosphere will be presented in a separate publication.

G. Results for Power Law Size Distributions

In Fig. 19–21, we present a few selected results for the case of a power law size distribution. Figures 19 and 20 show a series of error contours in \( \theta - \nu \) plane for refractive indices of 1.33–i0.0 and 1.55–i0.05, respectively. These two refractive indices usually produced the largest and smallest errors (respectively) for a given \( \theta - \nu \) combination. Since an increase in \( \nu \) leads to a reduction in large particle content, the shape of these plots is inverted compared with those in Fig. 17.

Figure 21 shows plots of \( E \) vs \( \theta \) for a refractive index of 1.55–i0.05 and for five selected values of \( \nu \). The Rayleigh approximation results are included for comparison. (Again, symbols without crosses represent Mie results; those with crosses represent Rayleigh results.) The shape of these curves is similar to those in Figs. 11–16. We see from Fig. 21 that the Rayleigh approximation is good for this complex refractive index over the full range of \( \nu \) values considered. As in the case of the Deirmendjian models, the Rayleigh approximation is not as good for a real refractive index.

VIII. Summary Remarks

From the foregoing discussion, it is obvious that the transmission law as expressed by Eq. (1) cannot be used except in the case of small particles, for which it is merely a good approximation. The effect of not only the particle size distribution, refractive index, and shape, but also the geometry of the optical system on the apparent volume scattering and extinction coefficients must be considered.

This study shows that the \( \overline{R(E)} \) decreases (increases) as the size distribution becomes more polydisperse. This result can be explained by the fact that as the polydispersity becomes greater, the relative proportion of larger particles increases, resulting in an increase in forwardscattered radiation within narrower angles. The results are valid for conditions in which multiple-scattering effects can be neglected, and only single scattering predominates.

The lens–pinhole detector geometry yields the most accurate transmission/extinction results, provided the design conforms to the prescription that the value of \( y \) in Fig. 18 should be such that \( R \) is close to either 1.0 or 0.5.

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References

REFERENCES


Table 1. Summary table for size distribution models.

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<th>Model 2</th>
<th>Model 3</th>
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<td>Power Law</td>
<td>Regularized Power Law</td>
<td>Modified Gamma</td>
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<td>n(r)</td>
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<td>$\frac{P_1}{P_2} \frac{(r/r_0)^{P_2-1}}{(1+(r/r_0)^{P_2-1})}$</td>
<td>$P_2 \exp(-P_1 r^{P_4})$</td>
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<td>S_n</td>
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<td>$\frac{P_3}{P_2} \left( \frac{P_3}{P_4} \frac{1}{(1+(r/r_0)^{P_3-1})} \right)$</td>
<td>$P_4 \exp(-P_3 r^{P_4})$</td>
</tr>
<tr>
<td>n(r)</td>
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<td>$\frac{P_2}{P_3} \left( \frac{1}{1+(r_0/r)^{P_2-1}} \right)$</td>
<td>$P_3 \exp(-P_2 r^{P_2})$</td>
</tr>
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</table>

As $r \to 0$, $n(r)$

As $r \to \infty$, $n(r)$

As $n(r) = n(r)$

As $r = 0$, $N(r)$

As $r = \infty$, $N(r)$

As $r = 0$, $n(r)$

As $r = \infty$, $n(r)$

As $n(r) = n(r)$

As $n(r) = n(r)$

$N_a = \frac{P_1}{(P_2 r^{P_2-1})} \left( \frac{1+k-P_2}{r_2} \right)^{P_1} \left( \frac{r^{P_2-1}}{r_0^{P_2-1}} \right)^{P_1} \left( \frac{r^{P_2-1}}{r_0^{P_2-1}} \right)^{P_4} \left( \frac{r^{P_2-1}}{r_0^{P_2-1}} \right)^{P_2}$
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<td>( 0 )</td>
</tr>
<tr>
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<td>( \frac{P_4}{\gamma_{d2}} \exp(-\frac{1}{2}(\frac{P_4}{P_2})^2 \frac{\exp(-\frac{1}{2}(\frac{P_4}{P_2})^2)}{P_2} )</td>
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<td>( r = \infty )</td>
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<td><strong>Normal Distribution</strong></td>
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(continued)
Table 1. Summary table for size distribution models (concluded).

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<td>( \frac{P_1 \exp(p_3/r)}{r^{p_4/1 + p_4 \exp(p_3/r - 1)^2}} )</td>
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<td>( \Gamma_n )</td>
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<td>See Eq. (11c)</td>
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<td>( P_1 r )</td>
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<td>( N(r) )</td>
<td></td>
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<tr>
<td>( p_k )</td>
<td>N.A.</td>
<td>N.A.</td>
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<td>( P_{kr} )</td>
<td>( P_{kr} (1 + 1) )</td>
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Figure 1. Relative intensities of sky radiance and Sun's direct radiation; $r_s$ represents distance from the Sun in solar radii (from ref. 14).
Figure 2. Geometry of the single scattering problem.
Figure 3. Schematic illustration of the solar aureole photographic equipment.
Figure 4. (a) A typical solar aureole 35-mm photograph  
take through a wavelength filter, $\lambda = 50$ nm.  
(b) An isodensity tracing of the photograph.  
(c) The computed solar isophote mapping of  
the photograph.
Figure 5. Almucantar projection on the film as a conic.
Figure 6. The shapes of the conics for various solar zenith angles $\phi$, for lens focal lengths of 35 mm and 55 mm.
Figure 7. Comparison of almucantar radiance measured by the photographic and the photoelectric methods.
Figure 8. Typical size distribution curves for parameter $\nu = 4.0$, 4.4, and 5.2.

Figure 9. A set of phase function curves corresponding to the size distribution curves shown in figure 8 for $\nu$ values in the range of 4.0 to 5.2.
Figure 10. A typical example of inversion of solar aureole data to retrieve aerosol size distribution. (a) Solid lines denote the final steps to the experimental data denoted by the samples, and (b) plots represent the corresponding retrieved results.
Figure 11. Computer-generated SS circumsolar isophotes for different values of the size distribution parameter $\nu$: (a) 4.0, (b) 4.4, and (c) 5.0.
Figure 12. Error contours in x–θ plane for m = 1.50 and percentage errors of 1, 5, and 10 percent.
Figure 13. Aerosol extinction coefficient altitude profile.
Figure 14. Model 2 (regularized power law) for n(r).
Parameter set 2.1: $p_2$ variable, $p_3 = 2.0$, $p_4 = 3.0$. 