Determination of optical parameters of atmospheric particulates from ground-based polarimeter measurements

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SUMMARY

This paper describes the theoretical analysis that is required to infer, from polarimeter measurements of skylight, the size distribution, refractive index and abundance of particulates in the atmosphere. To illustrate the viability of the method, some data obtained at UCLA is analysed and the atmospheric parameters are derived. The explicit demonstration of the redundancy in the description of aerosol distributions suggests that radiation field measurements will not uniquely determine the modal radius of the size distribution. In spite of this non-uniqueness information useful to heat budget calculations can be derived.

1. INTRODUCTION

The problem of the transfer of electromagnetic radiation in a plane parallel multiply scattering molecular atmosphere was solved by Chandrasekhar (1950). Computations of his results have been tabulated by Sekera and his collaborators (Coulson, Dave and Sekera 1960). Subsequently, extensive measurements of skylight polarization displayed substantial deviations from Chandrasekhar’s predicted results. Sekera (1951, 1956, 1957), quite correctly, ascribed this to the presence of particles in the atmosphere.

Sekera’s last years of research were directed toward determining the sizes of particulates from polarization measurements (1967). This is the so-called ‘inverse problem’ in atmospheric optics, in which the incident and scattered beams are specified and the nature of the scatterers have to be inferred. The equations of transfer in a turbid atmosphere are, however, complicated and they cannot be easily inverted. The operational method adopted here is, therefore, to solve the generic direct problem, tabulating all the results and then matching measurements against these tables in order to infer the sizes of particulates.

The efficiency with which the results can be matched against the tables will depend crucially on the length of the tables and the method of search adopted. Analytical work that was performed earlier (Kuriyan and Sekera 1972, 1974) led us to eliminate many redundant parameters and thus minimized the size of the required tables without any sacrifice in generality. The parameters of the problem were varied and the consequent changes in predicted intensity and polarization of skylight were studied using numerical methods. Such parametric analysis helped in the search for a fit to experimental data. This is a viable method but does not, as it stands, render itself well to computerized search techniques.

The paper is arranged as follows. In section 2 the radiative transfer equation in a turbid atmosphere and its formal solution are given. The phase matrix that appears in the solution is constructed in section 3. The sensitivity of the numerical solution that is obtained to the aerosol parameters is investigated in section 4. The apparatus and the experiment is described in section 5. The fitting of the data is accomplished in section 6 and the relevant aerosol optical parameters are tabulated. Section 7 concludes with a discussion of the implications of the results obtained, the limitations of the experiment and the outstanding problems that remain to be investigated.

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2. EQUATION OF TRANSFER IN A TURBID ATMOSPHERE

Following the notation of Sekera, the auxiliary equation for the source function \( J(\tau, \Omega) \) at an optical depth \( \tau \) and direction \( \Omega \) (which stands for the two parameters \( \mu \) and \( \phi \) where \( \cos^{-1} \mu = \text{zenith angle of observation} \), and \( \phi \) the azimuth angle of observation) due to the unpolarized radiation \( \pi F(0, -\Omega_0) \) incident on the top of the atmosphere (\( \tau = 0 \)) and in the direction \(-\Omega_0\), i.e. \((-\mu_0, \phi_0)\), when ground reflection is ignored, is given by

\[
J(\tau, \Omega) = 4e^{-\frac{\tau}{\mu_0}} \left\{ \sum \beta_i(\tau) P_i(\tau, \Omega, -\Omega_0) \right\} F(0, -\Omega_0)
\]

\[
+ \frac{1}{4\pi} \int_0^{2\pi} d\phi' \int_0^1 \frac{d\mu'}{\mu'} \left\{ \sum \beta_i(\tau) P_i(\tau, \Omega, +\Omega') \right\} \int_0^\tau e^{-\frac{\mu}{\mu_0}} J(t, +\Omega') dt
\]

\[
+ \frac{1}{4\pi} \int_0^{2\pi} d\phi' \int_{-1}^0 \frac{d\mu'}{\mu'} \left\{ \sum \beta_i(\tau) P_i(\tau, \Omega, -\Omega') \right\} \int_0^\tau e^{-\frac{\mu}{\mu_0}} J(t, -\Omega') dt
\]

(1)

with the definition

\[
\sum \beta_i(\tau) P_i(\tau, \Omega, \Omega') \equiv \frac{\beta_R(\tau) P_R(\tau, \Omega, \Omega') + \beta_A(\tau) P_A(\tau, \Omega, \Omega')}{\beta_R(\tau) + \beta_A(\tau)}
\]

(2)

where \( \beta_R(\tau), \beta_A(\tau) \) are the volume scattering coefficients; while \( P_R(\tau, \Omega, \Omega'), P_A(\tau, \Omega, \Omega') \) are the phase matrices for the Rayleigh and the aerosol parts, respectively.

From the source function the diffuse component of the intensity can be calculated. If the reflection at the ground is assumed Lambertian, another term, \( J^*(\tau, \mu) \), is added to the source function and the magnitude of this term is determined by \( A \), the reflectivity of the ground. The relevant expressions have been obtained by Dave (1964). We will borrow his results and focus our attention on the contribution that aerosols make to the transfer problem.

The phase matrix for aerosol scattering contains within it the dynamics of the interaction between the electromagnetic radiation and the particulates in the atmosphere. The object is to arrive at the simplest parameterization of the phase matrix that will be sufficiently general to include the diverse scattering processes that occur in the atmosphere. Naturally, this treatment is model dependent and the attempt is to derive the results of the basic model in such a way that our model calculations, even with the model dependency, can be used with sky polarization measurements to make relative statements of great value concerning the atmospheric aerosol constituents. We shall give an example of such an application in a later section.

3. CONSTRUCTION OF THE BASIC PHASE MATRIX

The phase matrix will depend on the physical properties of the scatterers such as radius, refractive index, abundance and so on and these will be related to the parameters of the model describing the turbid atmosphere. We shall consider a simple description of the polydispersion and then arrive at the appropriate phase matrix.

The particulates in the atmosphere are assumed to be spherical objects with varying radii and constant refractive index. They scatter incoherently according to the theory of Mie. The abundance of these particulates as a function of radius is called the size distribution function. Careful analysis of experimental data on atmospheric aerosols led Deirmendjian (1969) to use a modified gamma function to describe their size distributions,
\[ n(r) = ar^a e^{-br} \]  (3)

where, \( a, b, \alpha \) and \( \gamma \) are free parameters. The polydisperse phase function is then derived by integrating the scattering phase function of a single sphere (obtained from Mie theory), over all radii, with the size distribution as a weight function. Since the weight function and the Mie phase function are analytic it is possible to make the range of integration, \( r_{\text{min}} < r < r_{\text{max}} \), as large as we wish, an option that is not available with the alternative size distribution due to Junge (1963).

Deirmendjian considered three haze models: haze H (\( \gamma = 1, \alpha = 2 \)), haze L (\( \gamma = \frac{1}{2}, \alpha = 2 \)) and haze M (\( \gamma = \frac{1}{2}, \alpha = 1 \)), to characterize the typical particulate distributions of the stratospheric, continental and marine haze, respectively. The analytic formula that Kuriyan and Sekera (1972, 1974) derived indicated that, in the Born approximation, the haze H model duplicated the results of haze L and M when the parameter \( b \) was chosen appropriately. It was not clear at that time whether this redundancy existed in the Mie formulation as well. Subsequent work has confirmed that this is the case. Explicit examples of this will be provided in section 7. The remarkable agreement indicates that as far as the phase functions are concerned, the models H, L and M are redundant and thus the irreducible set of models need only include haze H. We shall exploit this equivalence by considering, at all times, a haze H distribution function and, after results are matched, the equivalent haze L and M distributions will be obtained.

In summary, the condition of analytic behaviour of the size distribution function led us to Deirmendjian's parameterization. The analytic approximation to the phase function raised the suspicion of a redundancy in Deirmendjian's description, which was confirmed by exact calculations. Thus we arrive at the basic or irreducible set of parameters \( a \) and \( b \) and the size distribution function:

\[ n(r) = ar^a e^{-br} \]  (4)

which is sufficiently general (at least to the extent that it includes haze H, L and M) to handle the numerous scattering processes that are of interest.

4. SENSITIVITY OF RESULTS TO VARIATION OF THE PARAMETERS

A numerical solution for the transfer problem in a turbid atmosphere was obtained by Herman and Browning (1965) and Dave (1970a, 1970b, 1970 (with Gazdag), 1972). This programme is capable of handling all possible vertical inhomogeneities and various size distributions. The programme has been used by Dave and collaborators (Braslau 1973) to make several useful calculations. We shall use Dave's programme (1972), with the simplification obtained in the earlier section (i.e. using Deirmendjian's haze H with a variable parameter \( b \)) to catalogue, exhaustively, all relevant results.

It is our aim to compare measurements against these tables and thus infer the physical characteristics of the scatterers. In order to facilitate this comparison it is necessary to identify the sensitivity of the results to variations in parameters such as \( b \), the refractive index, turbidity and so on. Since analytic studies are unavailable we must rely on representative numerical runs of the Dave programme to arrive at the relevant conclusions. We will consider only the downward radiation at the ground level. The experimental arrangement has the sensor scanning the sky at a fixed zenith angle. Thus our analysis will concern itself with azimuthal variations of the sensor for fixed zenith angles of the sun and the sensor.

Absolute intensity measurements contain important information on the optical thickness or the particle loading in the atmosphere. Unfortunately, these measurements require precise calibrations that are not too easy to perform. We shall consider intensities normalized
with respect to the intensity at zero azimuth (i.e. when the scattering angle has the smallest value) and side-step this question of calibration. The method we adopt will also derive the particle loading in the atmosphere.

The effects of variations in the aerosol model parameters on the computed scattered intensity \( I \) and degree of polarization \( P \) are shown in Figs. 1–4. The \( I \) and \( P \) curves for azimuth angles \( 0 \leq \Phi \leq 180^\circ \) are given in Fig. 1 for \( b = 10 \) and \( b = 25 \). Analysis of many such curves led us to the conclusion that \( b \) is a factor that determines the half-width of the bell-shaped \( I \) curve. That is, when \( b \) increases the \( I \) curve broadens. In Fig. 2 we see that the effect of increasing the Mie optical thickness \( \tau_m \) is to depolarize the radiation while the intensity curve retains its form. Fig. 3 shows that the increase of refractive index depolarizes. The tail of the intensity curve moves up as the refractive index increases. The effect of Lambertian albedo variation is shown in Fig. 4. Increasing the albedo causes depolarization and an increase in scattered intensity. Thus we have isolated the consequences of the variation of the parameters which will enable us to search for a fit to the experimental data.

5. EXPERIMENTAL APPARATUS AND METHOD

A prototype polarimeter was designed and manufactured for skylight measurements by the TRW Systems Group, Redondo Beach, Ca., and loaned to UCLA to conduct these
Figure 3. Normalized scattered intensity ($I$) and degree of polarization ($P$) for aerosol indices of refraction $n = 1.34$ and 1.54 as a function of azimuth angle ($\Phi$) relative to the solar vertical plane.

experiments. Typically, 0.5° and 0.2° fields-of-view are employed for low and high intensity scattering, respectively. The four observation wavelengths used in the instrument are $\lambda = 0.448 \mu m$, 0.575 $\mu m$, 0.701 $\mu m$ and 0.822 $\mu m$. Filter bandwidths are about 0.01 $\mu m$.

The quantities of interest that are derived from the measurements are the degree of polarization and the intensity of the scattered skylight. At present, only relative intensity measurements are made and the reported values are normalized to that of the solar vertical plane. The polarimeter is mounted on an alt-azimuth tracker. The tracker has angular readout scales in both azimuth and elevation, facilitating the precise determination of polarimeter orientation. The first and last measurements are made in the solar vertical plane and all other measurements are referenced in azimuth to this plane. In addition, the solar zenith angle and time of measurement are recorded. Sets of data are usually taken at observation zenith angles of 30°, 40°, 50° and 60°.

Figure 4. Normalized scattered intensity ($I$) and degree of polarization ($P$) for various Lambertian ground reflectances $A = 0$, 0.2 and 0.4 as a function of azimuth angle ($\Theta$) relative to the solar vertical plane.

The process of data reduction begins with computation of the solar zenith angles during the observations, using the times of the measurements, the values of solar declination, ephemeris transit and their diurnal variations. Any differences between observed and calculated solar zenith angles due to instrument misalignment can be used to correct the observation zenith angles as well. After correcting the data for instrument offset and drift, the degree of polarization and total, normalized intensity are computed.
6. FITTING OF THE DATA

The data were gathered on 12 August 1973 at UCLA using the TRW polarimeter. The measured values of scattered intensity \( I \) and degree of polarization \( P \) were plotted. Since \( b \) determines the width of the normalized \( I \) curve a match of the data against the compiled tables readily yields the value of \( b \). A reasonable value of reflectivity (Kondratyev 1973) was assumed (for \( \lambda = 0.7 \) we had \( A = 0.2 \)) and then the refractive index and the optical thickness varied till the \( I \) and \( P \) curves obtained from theory matched the data points.

![Figure 5. The solid curves are the computed \( I \) and \( P \) for the aerosol model whose parameters are: \( b = 25 \mu m^{-1} \), refractive index = 1.54, Mie optical thickness = 0.25 and ground reflectance = 0.2. The dots are measured values of \( I \) and \( P \) made at UCLA on 12 Aug. 1973 at 1425 hr Pacific Daylight Time. The wavelength for this observation was 0.701 \( \mu m \). The zenith angle of observation was 62.8° and the solar zenith angle 27.8°.]

These fits are displayed in Fig. 5, 6, 7. The albedo of the ground was, so to say, calibrated by the first measurement and further measurements studied the temporal variation of refractive index \( m \), optical thickness \( \tau \) and the modal radius \( r_c = 2/b \) of the size distribution. The parameters obtained by this fitting are given in Table 1. The corresponding size distribution functions are displayed in Fig. 8.

<table>
<thead>
<tr>
<th>TABLE 1. OPTICAL PARAMETERS FROM BEST FIT OF DATA</th>
</tr>
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<tbody>
<tr>
<td>Data set no.</td>
</tr>
<tr>
<td>--------------</td>
</tr>
<tr>
<td>1</td>
</tr>
<tr>
<td>2</td>
</tr>
<tr>
<td>3</td>
</tr>
</tbody>
</table>

\* Pacific Daylight Time.

\dagger \( \tau_{total} = \tau_{Rayleigh} + \tau_M \) and \( \tau_{Rayleigh} = 0.037 \) for \( \lambda = 0.7 \).

The uncertainties in the determination of the aerosol model parameters \( b, m \) and \( \tau \) can be ascribed to two principal sources of error: (i) random uncertainties arising from the polarization and intensity measurements; and (ii) the lack of perfect agreement between the observed \( I \) and \( P \) curves and the best fitting theoretical ones. We have analysed these sources of error and the sensitivity of our deduced \( b, m \) and \( \tau \) to them. While the exact uncertainties differ from one data set to the next, because of different measured voltage
levels and different degrees of success in fitting theoretical $I$ and $P$ curves to experimental ones, a general statement on errors can be made. For the three characteristic data sets discussed here $b$ can be determined to within $\pm 2 \mu m^{-1}$, the index of refraction ($n$) to within $\pm 0.02$ and the optical thickness ($\tau$) to within $\pm 0.02$.

7. DISCUSSION

In section 3 we had stated, without proof, that Deirmendjian’s haze $H$ with a variable $b$ was equivalent to haze $L$ and haze $M$. We shall demonstrate this equivalence for one set of parameters obtained from fitting the data, $b = 18$ and $m = 1.44$. In Figs. 9, 10 and 11, we have shown the computed, normalized phase matrix elements $P_1$, $P_2$, $P_3$ as functions of the scattering angle $\psi$, for haze $L$ with the parameters ($\alpha = 2$, $\gamma = \frac{1}{2}$, $b_L = 23$) and for haze $M$ with the parameters ($\alpha = 1$, $\gamma = \frac{1}{2}$, $b_M = 20$). These are compared with the corresponding haze $H$ ($\alpha = 2$, $\gamma = 1$, $b = 18$) results (indicated by dots). The index of refraction was 1.44 in all cases. The equivalence of the phase matrix elements assures us

Figure 6. The solid curves are the computed $I$ and $P$ for the aerosol model whose parameters are: $b = 12.857 \mu m^{-1}$, refractive index = 1.54, aerosol optical thickness = 0.12 and ground reflectance = 0.2. The dots are measured values of $I$ and $P$ made at UCLA on 12 August 1973 at 1438 hr Pacific Daylight Time. The wavelength for this observation was 0.701 $\mu m$. The solar and observation zenith angles were 28° and 41°, respectively.

Figure 7. The solid curves are computed values of $I$ and $P$ for the aerosol model whose parameters are: $b = 18 \mu m^{-1}$, refractive index = 1.44, aerosol optical thickness = 0.25 and ground reflectance = 0.2. The dots are measured values of $I$ and $P$ made at UCLA on 12 August 1973 at 1457 hr. Pacific Daylight Time. The solar and observation zenith angles were 33° and 62°, respectively.
that these different size distributions give rise to the same scattered radiation field. In Table 2 we have displayed the equivalent parameters for all three data sets and one set of size distribution functions are displayed in Fig. 12. Thus we have shown that radiation field measurements cannot distinguish the three types of haze distributions from one another. This non-uniqueness is confined solely to the parameter $b$.

Our numerical studies have shown that the equivalence between H, L and M persists for all reasonable values of the parameter $b$. Unfortunately, we have not arrived at a general proof for the necessary and sufficient conditions for such equivalence. We suspect that this equivalence exists because the phase function is determined not by $n(r)$ but a weighted integral of it, and in the region where the integrand makes a significant contribution $e^{-2\sqrt{r}}$ can be simulated by $e^{-b}$. Thus a haze H distribution yields a result similar to that of haze L or M.

**Table 2. Equivalent Optical Parameters**

<table>
<thead>
<tr>
<th>Data set no.</th>
<th>Haze H $(\alpha = 2, \gamma = 1)$</th>
<th>Haze L $(\alpha = 2, \gamma = 0.5)$</th>
<th>Haze M $(\alpha = 1, \gamma = 0.5)$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$b$</td>
<td>$b_L$</td>
<td>$b_M$</td>
</tr>
<tr>
<td>1</td>
<td>25</td>
<td>28</td>
<td>25</td>
</tr>
<tr>
<td>2</td>
<td>12.857</td>
<td>19</td>
<td>16</td>
</tr>
<tr>
<td>3</td>
<td>18</td>
<td>23</td>
<td>20</td>
</tr>
</tbody>
</table>

Despite this lack of uniqueness, we can make many useful relative statements. For instance, the modal radius $r_c = 2/b$ seems to change dramatically in our measurements. Presumably, the changing wind patterns alter the size distribution of aerosols in the coastal Los Angeles basin and this is manifested by a corresponding change in the 'average' radius. We observe from Table 2 and Figs. 11 and 12 that when the modal radius of haze H decreases (increases) the modal radius for the equivalent haze L or M also decreases (increases). Thus if we choose any one distribution as a basic model the relative change in a parameter can be determined without ambiguity.

Yamamoto and Tanaka (1972) have calculated the radiative effects of aerosols for representative values of the aerosol optical parameters. They conclude that the effect of
the interaction of the visible solar radiation with aerosols is comparable in magnitude with that of the solar infrared absorption in the atmosphere. Their results seem to suggest that optical depth and refractive index determine the heating effects and not \( b \). A precise calculation of the radiative effects is thus possible if the aerosol parameters are known. A global network of ground-based polarimeters can yield this information. In view of the non-uniqueness in the determination of \( b \) it may be necessary to compute the radiative effects for each of the derived distributions. Since the determination of \( \tau \) and \( m \) do not have any ambiguities it should not come as a great surprise if the radiative effects for the three distributions coincide.*

This paper has addressed itself to the question of interpretation of polarimetric measurements in a turbid atmosphere. Since clouds are often parameterized using a modified gamma distribution (Deirmendjian 1969; Hansen 1969) it is tempting to assert that the method can be generalized to obtain the size distribution in clouds. It must, however, be kept in mind that the assumption of horizontal homogeneity (implicit in the plane parallel approximation) may be valid only in the case of widespread and uniform cloud formations. Further, the optical thickness may be sufficiently large to require a more desirable scheme than the

* Note added in proof: Recent calculations indicate that the three equivalent distributions give rise to the same radiative effects. Thus the measured optical parameters can be used to calculate heating effects without ambiguity.
Figure 10. Normalized phase matrix element $P_2$ as a function of the scattering angle $\psi$. The upper solid curve corresponds to haze L ($a = 2, \gamma = \frac{1}{2}, b_L = 22$) and the lower one to haze M ($a = 1, \gamma = \frac{1}{2}, b_M = 20$). The dots are for haze H ($a = 2, \gamma = 1, b = 18$).

iterative approach adopted here. On the experimental side we have observed that polari-
meter measurements fluctuate rapidly when wisps of clouds pass in the field of view. While it may be safe to speculate that this device will detect the presence or absence of
clouds, quantitative statements will require further systematic observations and theoretical
analysis.

Our measurements indicate that a large number of particles in the submicron range
would escape detection if the usual in situ techniques (Junge 1963) were used. This point
deserves further scrutiny.

In our analysis we have used a real index of refraction, perhaps not an unrealistic
assumption in the visible region. The computer programme is capable of handling a complex
index of refraction but the compilation of a catalogue which includes a range of complex
values of $m$ will be quite expensive.

The programme allows for the inclusion of an arbitrary vertical profile for aerosols. In our analysis we have chosen Elterman's (1968) profile. It would, of course, be desirable
to investigate, systematically, the effect of the vertical profile on the radiation field measure-
ments. In the absence of a canonical procedure the vertical profile can be varied in an infinite
number of ways and the analysis will become an enormous task. This merits careful investi-
gation.
Figure 11. Normalized phase matrix element $\bar{P}_3$ as a function of the scattering angle $\psi$. The upper solid curve corresponds to haze L ($\alpha = \frac{2}{3}, \gamma = \frac{1}{3}, b_L = 23$) and the lower one to haze M ($\alpha = 1, \gamma = \frac{1}{3}, b_M = 20$). The dots are for haze H ($\alpha = 2, \gamma = 1, b = 18$).

Figure 12. Normalized size distribution functions for haze H and the equivalent hazes L and M corresponding to data set #1.
The late Professor Sekera (1967) suggested the measurement of upwelling radiation from a spacecraft to monitor atmospheric particulates. The aircraft measurements undertaken by him at UCLA ended inconclusively. Rao et al. (1973) made balloon measurements of the degree of polarization and they inferred the atmospheric particulate characteristics. In their discussion aerosol particulates were considered to be homogeneously mixed and aerosol scattering assumed not to produce any polarization. An experiment using a helicopter-borne polarimeter was conducted jointly by TRW Systems Group and UCLA (J. G. K.) for NASA Langley Research Center. The Dave Programme (with the inhomogeneously distributed Mie scattering aerosols) was used to infer the optical properties. By a judicious choice of observation angles it was possible to prevent the ground reflection from interfering with the interpretation of data. While it was possible to obtain the relevant numbers for two flights, extensive measurements are required and statistically significant amounts of data must be reduced before any definitive statements on this mode of operation can be made.

ACKNOWLEDGMENTS

The last years of Professor Z. Sekera were directed towards the remote measurements of atmospheric optical parameters. This work is an outgrowth of the research performed by one of the authors (J. G. K.) under Professor Z. Sekera and is, therefore, dedicated to his memory.

We are most grateful to Mr. A. Sabroff, Mr. L. Speltz and Mr. P. G. White of TRW Systems Redondo Beach for the construction and loan of their polarimeter for our research use. The assistance that Mr. K. Jenkin of TRW rendered during the course of the experiment is also acknowledged. The NASA Computer programme was made available through the kind efforts of Dr. R. S. Fraser and Dr. M. P. McCormick. The invaluable expertise of Mr. Dennis St. John enabled us to modify, in a very short time, this huge computer programme to meet our specific needs.

We are especially thankful to Professor S. V. Venkateswaran for his valuable support of this experiment.

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