The Inversion of Aureole Measurements to Derive Aerosol Size Distributions

J. T. TWITTY

Dept. of Meteorology, University of Wisconsin Madison 53706

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ABSTRACT

An iterative method to invert size distributions from simulated scattered radiance measurements at small angles from the sun has been investigated. The inferred size distributions were represented by piecewise linear and cubic spline functions. Various relevant characteristics were investigated and it was found that:

1) The inverted size distribution was insensitive to the number of knots in the piecewise linear spline.
2) Within the range of sensitivity, the choice of initial guess had little effect on the inverted size distribution.
3) Five percent random noise in the simulated radiances appreciably deteriorated the result but variations are still tolerable when compared with other methods for determining size distributions.
4) The inverted distribution was insensitive to the index of refraction used in the kernel for particle radii $r > 1$ μm.
5) The choice of wavelength between 0.40 μm and 0.70 μm has negligible effect on the inverted distribution.
6) A range of tropospheric aerosol size distributions give acceptable inverted results.
7) The cubic spline representation can give reasonable inverted distributions, but may become unstable.

1. Introduction

The diffraction of sunlight by aerosols in the atmosphere produces a bright zone within a small angular zone around the sun. This effect is known as the solar aureole. The shape of this forward-scattered radiance is primarily a function of the size of the aerosols. It will be shown that aerosol size distributions can be derived from the diffuse radiance of the sky around the sun at angles out to 20°. This paper will demonstrate, for various tropospheric aerosols, the effects on the inferred size distributions of 1) the numerical procedure, 2) a lack of prior knowledge regarding the composition of the aerosols, 3) simulated noise in the radiances, and 4) the choice of wavelengths of the radiances.

2. Statement of problem

a. Definitions

Information regarding the aerosol size distribution is contained in the phase function $\bar{P}(\theta)/4\pi$ which characterizes radiance scattered at the angle θ (see Deirmendjian, 1969):

$$\bar{P}(\theta)/4\pi=\lambda^2\int_0^\infty K(r,\theta)N(r)dr/4\pi^2\beta_{\text{scat}}, \tag{1}$$

where λ is the wavelength of the scattered radiance, β_{scat} the scattering coefficient, $K(r,\theta)$ the Mie scattering kernel at a given wavelength which is a function of particle size and index of refraction of the material which comprises the aerosol, and $N(r)$ the size distribution of the aerosol. The aerosol is assumed to consist of spherical particles and to be distributed in horizontally homogeneous layers. For the purposes of this analysis, $\bar{P}/4\pi$ is the average over both polarizations. Since the direct solar beam is unpolarized and the polarization at near forward angles is small, there is no loss in generality produced by using this average.

The singly-scattered radiance $R(\theta)$ can be written in terms of this normalized phase function as

$$R(\theta)=\bar{P}(\theta)\tau F_0 e^{-\tau/\mu_0}/4\pi\mu_0, \tag{2}$$

where μ_0 is the cosine of the solar zenith angle, F_0 the extraterrestrial solar spectral irradiance at the wavelength being considered, and τ the aerosol scattering optical depth. The total scattering optical depth τ may be written

$$\tau=\int_0^\infty \beta_{\text{scat}}(z)dz, \tag{3}$$

where $\beta_{\text{scat}}(z)$ is the scattering coefficient and z the vertical height of the scattering layer. Eq. (2) holds only in the solar almucantar as long as multiply-scattered radiance can be ignored. [This is the problem treated here; the correction for multiple scattering is discussed in the previous paper by Weinman *et al.*

(1975)]. Since the scattering optical depth can be obtained from the direct solar extinction (assuming no absorption) and μ_0 and F_0 are known, then $\bar{P}(\theta_i)$ can be calculated.

Furthermore, only $K(r,\theta)$ is a function of θ, so it is convenient to write the inversion equation as

$$\bar{P}(\theta_i)=c\int_0^\infty K(r,\theta_i)N(r)dr, \qquad (4)$$

where c is a normalizing constant which scales the size distribution. The inverted solution yields only the relative size distributions. $\beta_{scat}(z)$ must be known if absolute number densities are to be obtained. The inverted distribution can be integrated to give $\beta_{scat}(z)$ if the vertical density function is known. For this analysis a thin uniform layer is assumed and the absolute distribution is then scaled by β_{scat}.

Eq. (4) is a Fredholm equation of the first kind. A solution consists of a determination of $N(r)$ from a set of values $\bar{P}(\theta_i)$, obtained from radiance values at angles θ_i.

b. *Information content*

Chahine (1972) points out that I measurements can yield no more than I independent values of $N(r)$. In general, the number of parameters of the function $N(r)$ that can be obtained in an inversion problem is less than the number of measurements. This is because the measurements are not independent. Twomey (1966) has shown that when an error term is considered in each measurement, it is possible to express some measurements within the described error as a linear combination of all the other measurements. This means that in any data set some of any set of measurements do not provide any additional information regarding the aerosol size distribution. Twomey and Howell (1967) have shown that forward-scattering measurements could yield at most five independent parameters.

Dave (1971) has shown that a set of overdetermined measurements improves the obtained inversion; thus, it is advantageous to measure the aureole at more than the minimum five angles. Unless the aerosol size distribution is known *a priori*, the information content of five measurements is uncertain. Redundancy increases the probability that a measurement will be taken at an angle where the information input to the size distribution is significant.

c. *Theoretical limits*

There are theoretical limits on the sensitivity of the Mie kernels to aerosol size. Fig. 1 provides qualitative insight regarding these limits. Each curve represents the kernel as a function of angle for selected values of the size parameter, $x=2\pi r/\lambda$. These kernels have been smoothed to remove the major ripples. An upper and a lower limit may be inferred in the following way. The curves for $x=125$ and $x=160$ are nearly identical. This

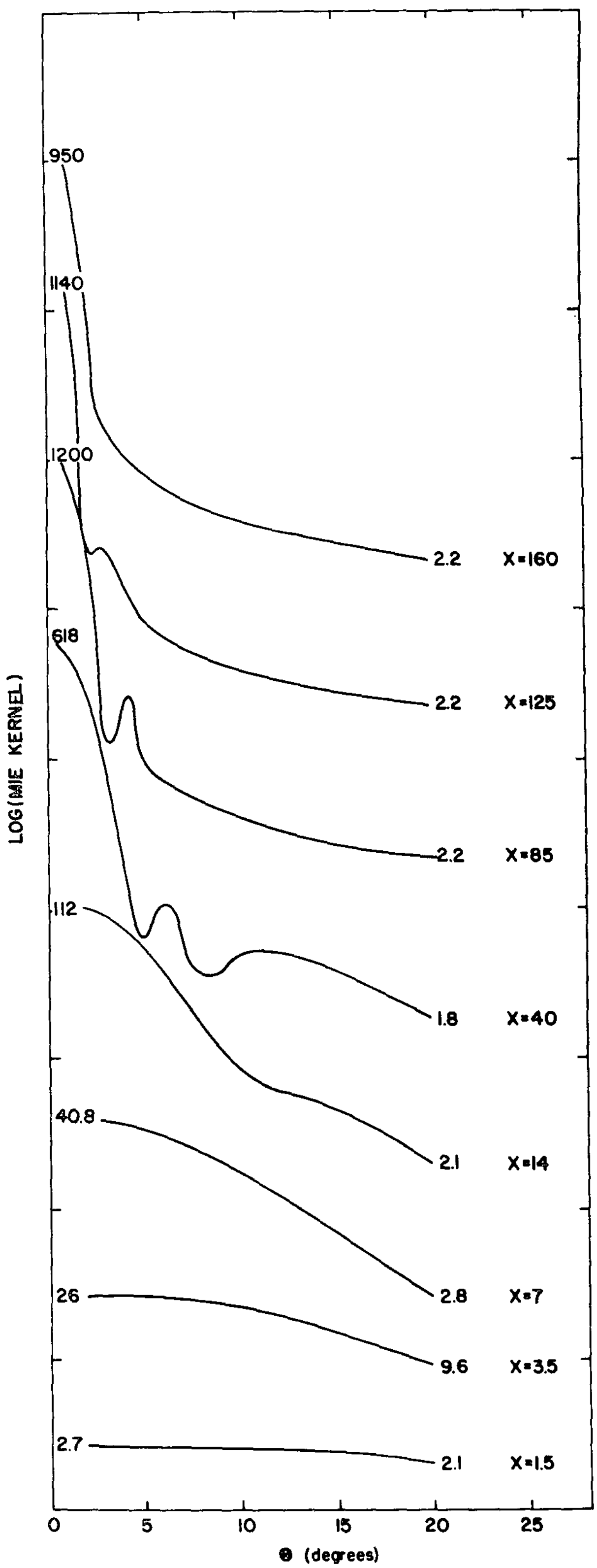

FIG. 1. Mean Mie kernels as a function of scattering angle for various values of the size parameter. For small size parameters the kernel is flat, while for large values the diffraction peak is less than 1° wide and the kernel remains relatively independent of size. Numbers on ordinate are the value of the kernel at 1° and 20°. Ordinate tic marks represent one decade.

is because the scattering function asymptotically approaches the geometrical optics limit. The diffraction peak in the forward direction for large x is much less than 1° wide and is diffused by the angular width of the solar disk. The inversion cannot distinguish between the relative number of particles of these sizes.

The lower limit is inferred from the $x=1.5$ curve. This is nearly flat and therefore cannot be distinguished from the Rayleigh case. Although not exact, one could set these theoretical limits at $3.0<x<100$. The relative shape of the size distribution can be obtained only between these limits. For the wavelength $\lambda=0.55\ \mu$m, this range corresponds to $0.25\ \mu\text{m}<r<8.0\ \mu$m. The inversion method may infer particles greater than these limits, but there is no further information as to their size. In addition, even within this range, if the number of particles is so small as to be optically negligible, then only an upper limit on the number of particles may be inferred.

3. Description of inversion method

a. General method

Chahine (1968) described an iterative method for inverting satellite radiance data to obtain temperature profiles. This technique has been modified to solve the problem at hand. The basic problem is to obtain the approximate size distribution function $N(r)$ from measured values of the function $\bar{P}(\theta_i)$ at various angles θ_i as defined in Eq. (4).

The steps in the general relaxation method of Chahine are as follows:

1. Guess an initial function $\underset{\sim}{N}^0(r)$.
2. Compute the corresponding measurements

$$\bar{P}^0(\theta_i)=c\int_0^\infty \underset{\sim}{K}(r,\theta_i)\underset{\sim}{N}^0(r)dr. \tag{5}$$

3. Make a new estimate of $\underset{\sim}{N}_i^j(r)$ for each measurement i by rescaling the previous estimate at the points r_k as follows:

$$\underset{\sim}{N}_i^j(r_k)=\underset{\sim}{N}^{j-1}(r_k)\bar{P}(\theta_i)/\bar{P}^{j-1}(\theta_i),\quad i=1,\ldots I. \tag{6}$$

4. Calculate an improved estimate by assuming that the estimates in (3) are independent and compute a weighted average. A recursion relation may be written between the jth and the $(j-1)$th estimate, i.e.,

$$\underset{\sim}{N}^j(r_k)=\sum_{i=1}^{I}\underset{\sim}{N}_i^{j-1}(r_k)W(r_k,\theta_i)/\sum_{i=1}^{I}W(r_k,\theta_i), \tag{7}$$

where $W(r_k,\theta_i)$ is the fractional contribution from the kth radius interval to the radiance at the angle θ_i. The subscript k has been added to the equation to indicate that the estimates are computed at descrete values of r_k.

5. Repeat (2)–(4) and set a convergence criterion based on the residuals D_j:

$$D_j=I^{-1}\{\sum_{i=1}^{I}\{[\bar{P}^j(\theta_i)-\bar{P}^0(\theta_i)]^2/\bar{P}^0(\theta_i)\}^2\}^{\frac{1}{2}}, \tag{8}$$

where j is the number of iterations considered. This is discussed further in regard to actual solutions.

6. Set $N_{\text{fit}}(r)=N^j(r)$ for the iteration which satisfies a specific convergence criterion. In the analysis presented here 100 iterations were preformed for all cases and no criterion was set.

b. Renormalization and interpolation

Aerosol size distributions frequently vary over the range of sensitivity of this inversion method such that $N(r)\propto r^{-\nu}$, where $\nu=3\pm1$. For small angle scattering the Mie kernel $K(r,\theta_i)\propto r^4$. This makes it convenient to redefine the above quantities so that less rapidly varying functions may be manipulated. The renormalized functions $\underset{\sim}{N}(r)=r^4N(r)$ and $\underset{\sim}{K}(r,\theta_i)=K(r,\theta_i)r^{-4}$ are used in this analysis.

At each iteration, the inversion procedure yields estimates of the size distribution at discrete points r_k. In addition each iteration requires estimates of $\underset{\sim}{N}(r)$ between the discrete points in order to evaluate the integral in (5). Two continuous estimates have been considered in this paper. The first is to let $\underset{\sim}{N}(r)$ be a piecewise linear spline. The second is to fit a cubic spline to the $\underset{\sim}{N}(r_k)$.

c. Spline functions

The underlying assumption in the use of spline functions is that the unknown function $\underset{\sim}{N}(r)$ can be well approximated by piecewise smooth polynomials. Greville (1969) describes the spline procedure in detail. One is given the value of the function $\underset{\sim}{N}(r_k)$ at the points $r_0,\ \ldots,\ r_k,\ r_K$. These points are known as knots. The approximation of $N(r)$ will be the function

$$\tilde{N}(r)=\alpha_{k0}+\alpha_{k1}(r-r_{k-1})+\alpha_{k2}(r-r_{k-1})^2+\alpha_{k3}(r-r_{k-1})^3, \tag{9}$$

where $k=1,\ \ldots,\ K$.

If a piecewise linear distribution is used to represent the size distribution, the coefficients α_{kl} are defined by the conditions (i) $\tilde{N}(r_k)=\underset{\sim}{N}(r_k)$, (ii) $\alpha_{k2}=\alpha_{k3}=0$ for all k, and (iii) $\tilde{N}(r)$ is a continuous function of r.

If a cubic spline is used to represent the size distribution, the coefficients α_{kl} are uniquely determined by (i) $\tilde{N}(r_k)=\underset{\sim}{N}(r_k)$, (ii) $\tilde{N}(r)$, $\tilde{N}'(r)$ and $\tilde{N}''(r)$ are continuous functions of r, and (iii) $\tilde{N}'''(r_0)=\tilde{N}'''(r_K)=0$. The first condition in both representations insures that the functions $\tilde{N}(r)$ are approximations of $\underset{\sim}{N}(r)$, since they take on the same values at the points where $\underset{\sim}{N}(r)$ is known. In the case of a cubic spline, the second condition insures that the approximate function is a smooth function. The third cubic spline condition is necessary to insure uniqueness of a spline interpolation.

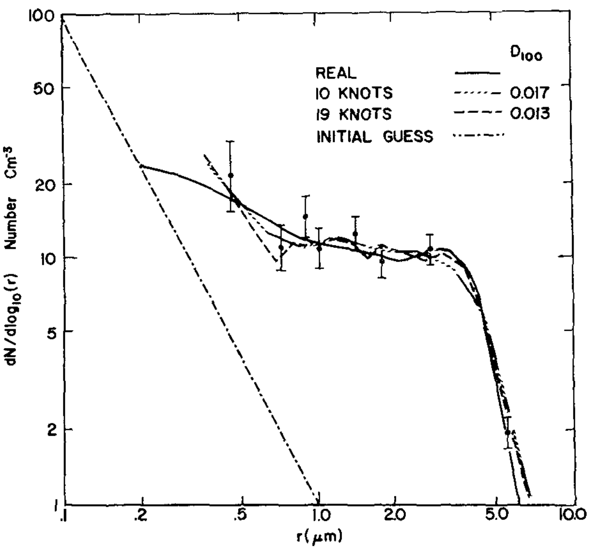

Fig. 2. The effect of increasing the number of knots used in the inversion. The error bars are for the impactor data. The fit to the data used to simulate the measured radiances and the inverted size distribution with 10 knots and with 19 knots are shown along with the initial guess.

The advantage of approximating the function by piecewise continuous functions is that the integral

$$\bar{P}(\theta_i)=c\int_0^\infty \underset{\sim}{K}(\theta_i,r)\tilde{N}(r)dr \qquad (10)$$

can be written

$$\bar{P}(\theta_i)=c\sum_{k=0}^{K}\sum_{l=0}^{3}\alpha_{kl}\int_{r_{k-1}}^{r_k}\underset{\sim}{K}(\theta_i,r)(r-r_{k-1})^l dr. \qquad (11)$$

This means that the quantities

$$\int_{r_{k-1}}^{r_k}\underset{\sim}{K}(\theta_i,r)(r-r_{k-1})^l dr \qquad (12)$$

are computed just once.

4. Sample distribution

This general relaxation method has been applied to simulated data to determine how well the inversion scheme may be expected to perform for solar aureole data. A size distribution obtained by impactor methods during a May 1972 field experiment was selected as a test distribution (Gillette, 1973, personal communication). This distribution is shown in Fig. 2. The error bars on typical impactor measurements may be 30% or more. Since size distributions vary over several orders of magnitude, errors this large are acceptable. A cubic spline function was fit to the data to generate a smooth curve, so that realistic simulated data could be calculated. Using the method of Reinsch (1967), the spline was allowed to differ from the actual data points, but remain within the error bars. A set of simulated radiances were generated from this spline fit assuming an index of refraction $m=1.54$.

In addition, three Deirmendjian (1969) haze models were used as a test of the inversion.

Except where noted, the following conditions were used in the inversion:

1. Radiances were measured at 20 angles, 1°(1°)20°.
2. Ten knots were located at $r_k=0.375$, 0.625, 0.825, 1.25, 1.75, 2.50, 3.50, 4.50, 5.50, and 6.50 μm.
3. Index of refraction, $m=1.54$. Absorption is not considered.
4. Initial guess $N^0(r)=1/r^3$.
5. Inverted size distribution approximated by piecewise linear function.
6. Wavelength of radiance was 0.54 μm.
7. 100 iterations were performed. The residual D_j decreased monotonically in all cases.

The residual from Eq. (8), D_{100}, is displayed for all cases as a measure of "goodness of fit." The function D_j is plotted as a function of the number of iterations in Fig. 3 for several typical cases; D_j tends to decrease asymptotically to a non-zero value. This tendency has been noted by Chahine (1972). For those cases with no noise added to the data, this represents the error inherent in the inversion and size distribution approximation. The poor fit obtained with the incorrect kernel (Fig. 4) indicates that the aureole has some sensitivity to the index of refraction which depends on

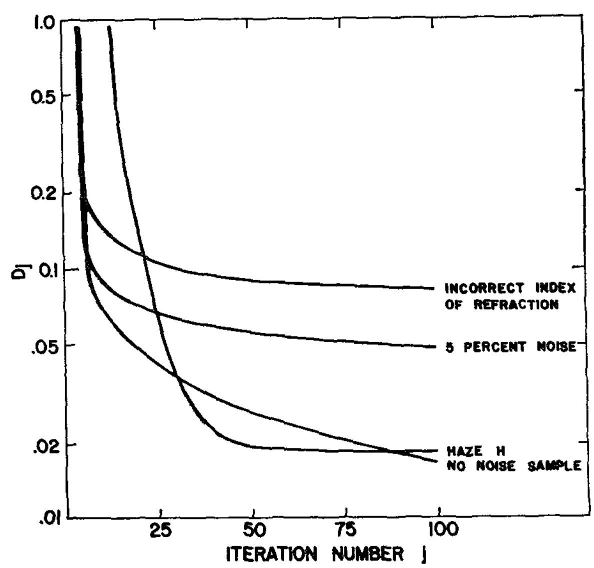

Fig. 3. Convergence of iterative method showing the rate of convergence of the estimated radiances to the actual radiances, D_j, as a function of the number of iterations j for various cases. The three similar curves had the same initial guess and actual distribution corresponding to Fig. 1. For haze H the convergence is at first slower, since the initial guess was a poorer estimate of this function.

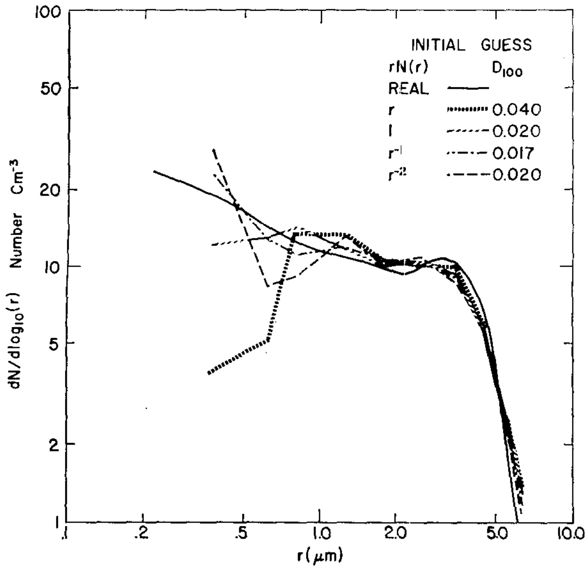

FIG. 4. The effect of the initial guess on the solution showing the resulting solutions for four initial guess power laws. The values of the residuals, D_{100}, are presented. The least acceptable fit also gives the largest residual.

the size distributions considered in the inversion procedure.

5. Summary of results

a. Number of knots

The influence that the number of knots had on the inversion was tested by comparing the inverted solution using both 10 and 19 knots. The 19 knots were located at r_k=0.375, 0.565, 0.675, 0.825, 0.950, 1.225, 1.375, 1.625, 1.875, 2.25, 2.75, 3.25, 3.75, 4.25, 4.75, 5.25, 5.75, 6.25, 6.75 μm. These curves are shown in Fig. 2.

Both inverted solutions are within the error bars of the original impactor measurements.

b. Initial guess effect

The effect of the initial guess on the result is shown in Fig. 4 for four initial guesses of the form $N(r)\propto r^{-\nu}$ where ν=0, 1, 2, 3. The results are again reasonable for r>0.6 μm. For r<0.6 μm the inverted distribution is dependent on the initial guess with poor initial guesses giving slower convergence. For the case with ν=0, the value of D_{100} is 0.040, twice as large as for the other cases. For most iterations, the number of small particles continues to increase very slowly. The choice of initial guess is not important for r>0.6 μm, but it affects convergence and the resulting distribution for smaller particles.

c. Effect of random noise

Eight independent samples with 5% noise added to the simulated measurements were generated. Although the deviations become greater when noise is added, the accuracy between r=0.5 μm and 6.0 μm is still comparable to that which direct measurements would provide. These results are shown in Fig. 5.

d. The effect of incorrect index of refraction

An estimate of the sensitivity of the size distribution to the choice of index of refraction in the Mie kernel was tested by changing the value of the index of refraction to 1.33 for the inverting kernel. The original measurements were obtained assuming an index of refraction m=1.54, corresponding to quartz, while the inverted kernel corresponds to water. This represents a typical range of tropospheric hazes. Fig. 6 displays the inverted distribution. It is evident that the incorrect index of refraction gives a poor distribution of aerosols with radii <1 μm because the small particles contribute more strongly to the large-angle scattering. This affect was later found to depend on the aerosol size distribution.

e. Wavelength dependence of inversion

Calculations have also been conducted to determine if the size distributions obtained by inverting aureole data are a function of the wavelength at which the measurements are taken. Data were simulated at wavelengths of 0.40, 0.54 and 0.70 μm. The inverted distributions shown in Fig. 7 were computed using the correct index of refraction and the same 20 angles. The differences in the inversion for the three different wavelengths is at most a few percent over the entire range of radii. There is little change in sensitivity over this

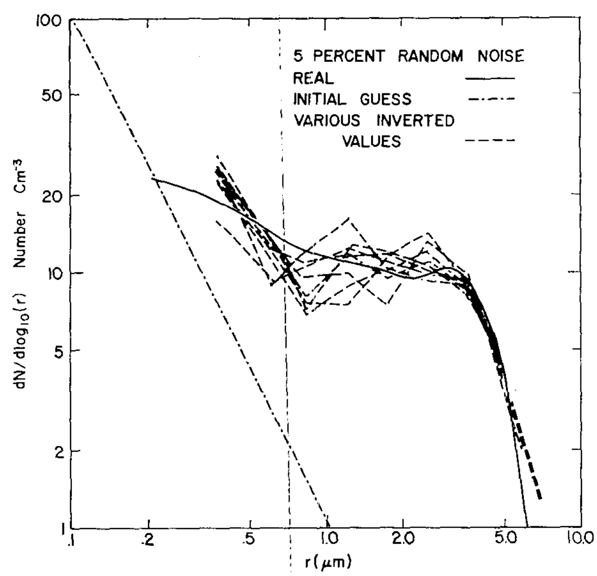

FIG. 5. The effect of 5% random noise added to the simulated data. Eight independent cases are displayed, giving the range of scatter induced by the noise. The value of D_{100} are also increased by the noise.

range of wavelength. For distributions which extend beyond the range of sensitivity of this inversion, additional wavelengths may add information to the solution, but for typical tropospheric hazes, a single visible wavelength appears sufficient.

f. *Dependence of inversion on aerosol size distributions*

Three other size distributions typical of tropospheric hazes were tested using the piecewise linear and third-order spline versions. Deirmendjian's (1969) water hazes M, L and H at a wavelength of 0.45 μm were used as input data. The inverted results are plotted in Fig. 8 together with the actual distributions. Contributions to any measurement between any two consecutive knots for sizes outside the plotted range were less than 2%. The inversion can, at most, establish that the number of particles outside the plotted range is optically negligible.

g. *Cubic spline variant of the inversion method*

The cubic spline variant of the inversion method was tested on all the cases above with similar results except when the size distribution has a sharp cutoff as in haze H. Although the cubic spline gives a continuous function, it is not constrained to positive values. If there is a sharp discontinuity in the distribution, the spline may go negative and yield an unrealistic solution. Fig. 8 also shows some spline results. For haze H the cubic spline procedure becomes unstable and drastic fluctuations are produced. The unacceptable solution gives a value of $D_{100}=0.365$.

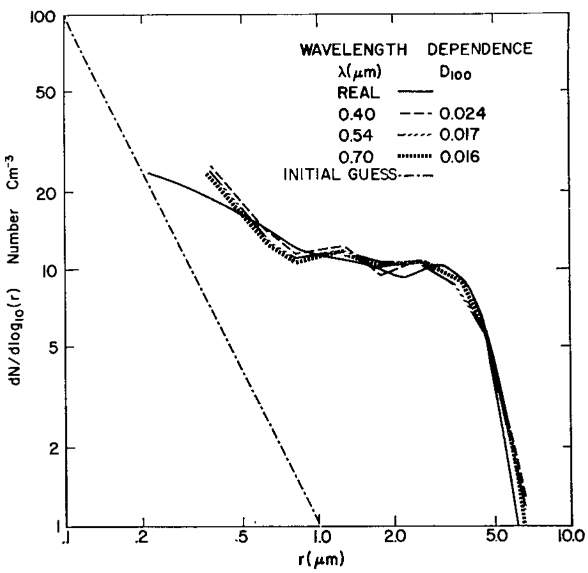

FIG. 7. Dependence of inverted size distribution on the wavelength of scattered radiance. Three different wavelengths (λ=0.40, 0.54, 0.70 μm) were used. There is little difference in the solutions, although the λ=0.40 μm case gives a slightly higher value of D_{100}.

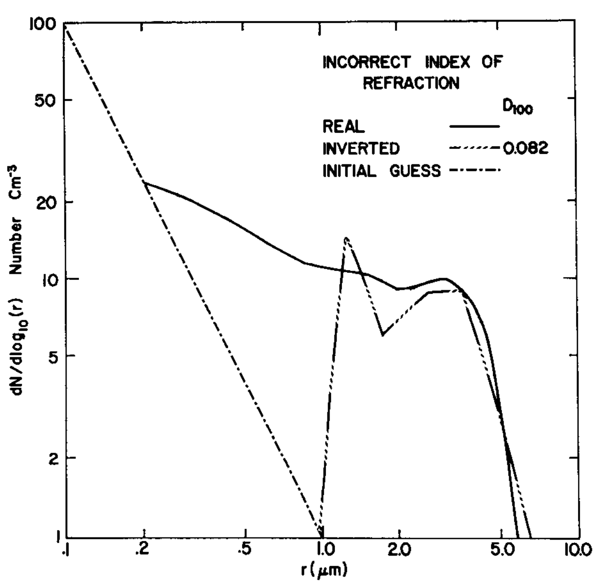

FIG. 6. The effect of incorrect index of refraction for inverting kernel on the inverted size distribution. Use of $m=1.33$ instead of $m=1.54$ causes poor results for $r<1.0$ μm. The value of D_{100} is also larger indicating a poor fit to the data.

6. Conclusions

Aerosol size distributions can be obtained from atmospheric scattering radiance measurements without *a priori* knowledge for distributions with radii in the range 0.25 μm$<r<$8.0 μm. This is the typical range of optically active tropospheric hazes.

The determination of size by the aureole method is as accurate as the direct method using impactors. While an impactor measurement is a localized determination at a point in space where the measurement was performed, the aureole determination is a mean over the optical path. Thus, the inversion is representative of the entire atmosphere above the instrument.

The solution to any inversion problem of the type considered here is not unique. Any meaningful solution must satisfy two conditions. First, physically unrealistic solutions must be eliminated from consideration. The general relaxation method used in this analysis damps out such high-frequency solutions by the averaging procedure. Second, small changes in input parameters must not induce intolerable differences in the solution. The results presented here indicate that this is the case for most of the size range of interest.

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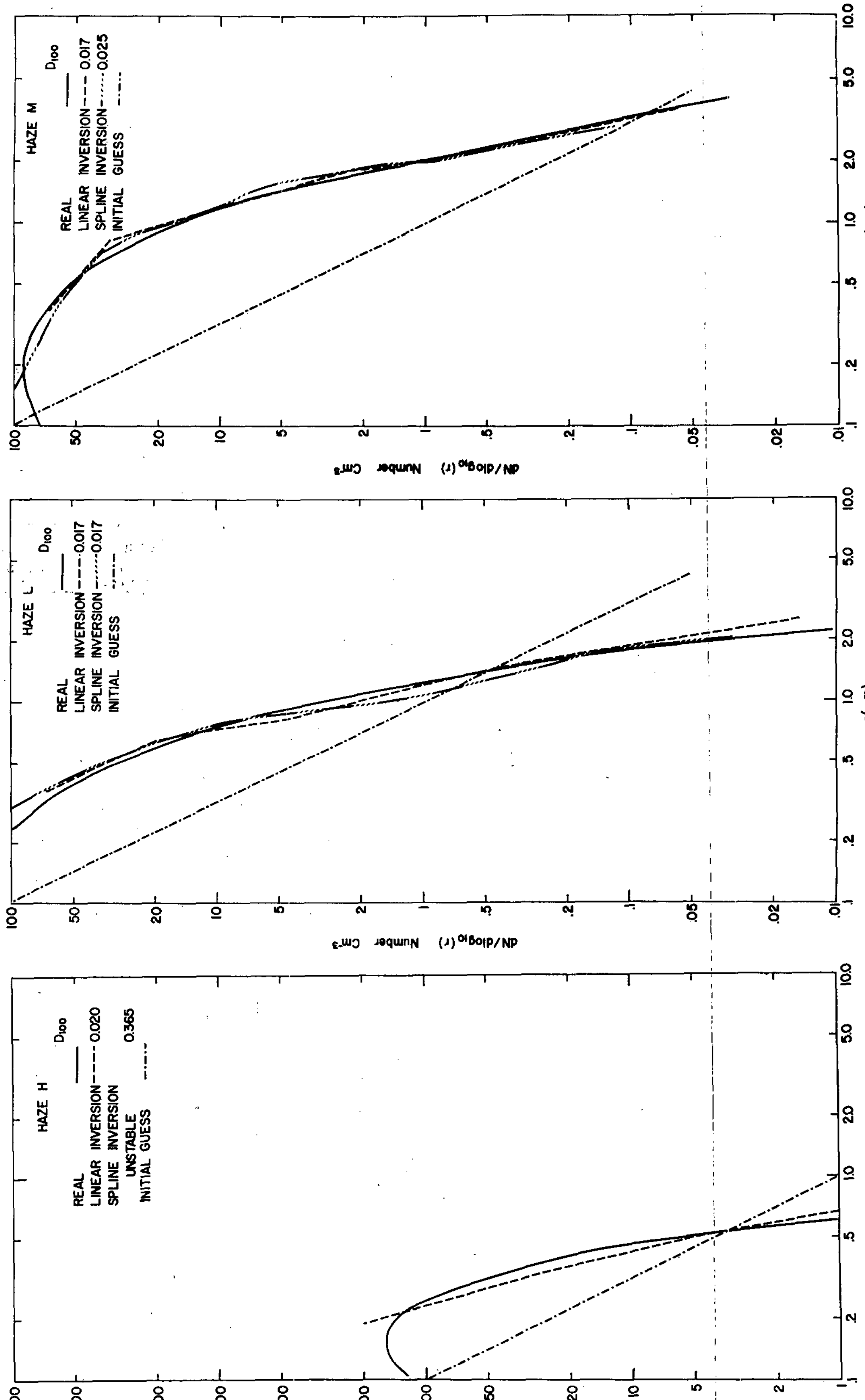

FIG. 8. Application of both linear and spline versions to three Diermendjian tropospheric water hazes. The cubic spline and linear cases give similar results for hazes M and L, but for haze H the cubic spline becomes unstable, giving a physically unrealistic solution.

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