Multi-Spectral Extinction Measurements to Deduce the Complex Refractive Index and the Size Distribution of Aerosol Particles

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ABSTRACT

It is shown that high-precision extinction measurements over a spectral interval can be used to infer aerosol optical parameters including the, heretofore elusive, complex index of refraction, if aerosol particles in the atmosphere are assumed to be spherical Mie scatterers with a uniform refractive index over this spectral interval and their sizes are distributed according to the modified gamma function. The error analysis shows that the degree of precision required is attainable, thus making it a viable and unique real-time remote sensing device. No assumptions are made on the vertical profile of aerosols and hence, in a satellite application (occultation experiment), the difficulties due to the sphericity of the medium can be avoided.

1. Introduction

The climatic effect of aerosol particles in the atmosphere has, of late, attracted a great deal of attention (e.g., Kondratyev, 1972). Reasonable calculations are lacking only because aerosol model parameters, such as refractive index, size distribution and abundance of particles have not been experimentally determined. The degree of precision and vastness of the area under investigation rule in favor of remote, rather than *in situ* techniques.

The polarimeter is one such passive remote sensing device and it was used successfully to infer some of the relevant parameters by Rao *et al.* (1973), Kuriyan (1974) and Kuriyan *et al.* (1974b). The bulk properties of the medium are described by these parameters and it was shown that this was by no means a unique description. However, anyone of the alternate descriptions gave rise to the same radiative effects. So, if the object were to estimate heating effects of aerosols, then the non-uniqueness is of no consequence and hence the term "equivalent parameterization."

The radiometer is another passive device designed to deduce the extinction optical thickness of a medium. Multi-spectral radiometric observations have been used by Yamamoto and Tanaka (1969) and Shaw *et al.* (1973) to yield the size distribution function as well. The thrust of this note will be to show that the refractive index, which has heretofore eluded measurement, can be determined for a specified size distribution.

2. Analysis

Deirmendjian (1969) has shown that the size distribution of naturally occurring aerosol particles can be represented by the modified gamma function. Kuriyan *et al.* (1974a, b) have shown that Deirmendjian's haze H, L and M are equivalent parameterizations and, so far as radiative effects are concerned, anyone of the descriptions would suffice. Here we choose the size distribution $n(r)=a(h)r^2e^{-br}$ and as-

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sume that the aerosol particles are spherical Mie scatterers with a uniform refractive index and varying radii. The extensive polarimeter observations conducted at UCLA by Kuriyan (1974) and Kuriyan *et al.* (1974b) suggest that this is entirely satisfactory and usual situations are accommodated if b is restricted to lie in the range $8 \leqslant b \leqslant 30$. An intrinsic advantage of Deirmendjian's distribution is its analytic behavior which obviates the need for a cutoff in the integral. In our analysis the size of particles is conveniently restricted to lie in the region $0.005\ \mu\text{m} < r < 6\ \mu\text{m}$ since the exponential damping assures that there is a negligible contribution for $r > 6\ \mu\text{m}$. The parameter $a(h)$ incorporates the vertical profile of aerosols and is proportional to the abundance of particles, i.e., particle loading. We do not have to assume the specific form of $a(h)$ since the results will apply to the integrated quantity $\int a(h)dh$.

The aerosol optical depth τ can be written as

$$\tau(h,\lambda,m) = H(h)\pi \int_{r_1}^{r_2} r^2 n(r) Q_{\text{ext}}(x,m) dr, \tag{1}$$

where $H(h)$ is the height dependent term which takes into account the vertical profile of aerosol distribution, $Q_{\text{ext}}(x,m)$ is van de Hulst's (1957) extinction efficiency factor, which is a function of the dimensionless size parameter $x \equiv 2\pi r/\lambda$ and the constant refractive index m of the particulates.

The consensus of Yamamoto and Tanaka (1974) and Bullrich *et al.* (1974) seems to be that for most polydispersions the real part of the refractive index varies between 1.34 (for water) and 1.54 (for sulphate particles). For urban areas with carbonaceous sources 1.5 seems a reasonable value. However, there is no convincing argument for the magnitude of the imaginary part of the refractive index. It is expected to be less than 0.1.

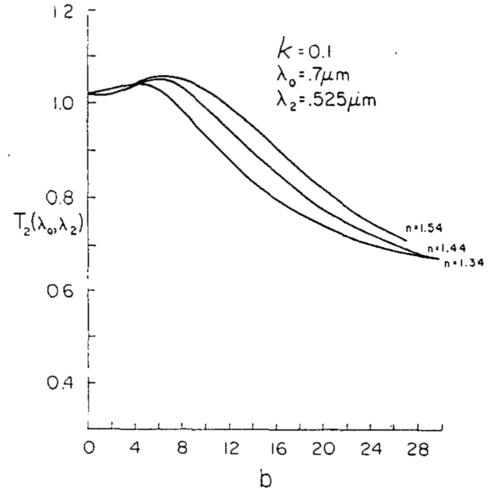

FIG. 1. The optical depth ratio $T_2(\lambda_0,\lambda_2)$ as a function of the aerosol parameters b and n, for $k=0.1$.

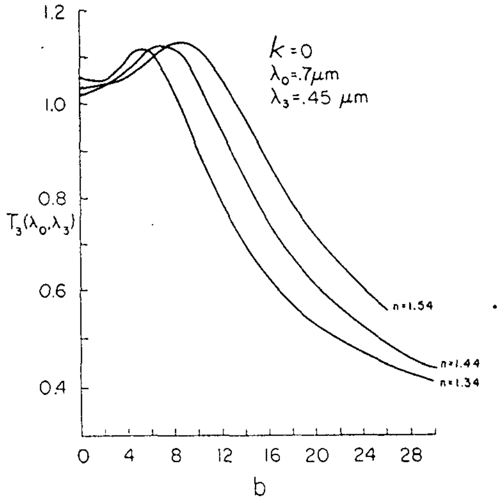

FIG. 2. The optical depth ratio $T_3(\lambda_0,\lambda_3)$ as a function of the optical parameters b and n, for $k=0$. Note the dependence of T_j on λ_j by comparing Figs. 1 and 2.

From multi-wavelength extinction measurements the aerosol optical depths $\tau(\lambda_j)$, $j=0, \ldots, J$, can be determined. Height dependence can be eliminated by considering only the ratios

$$T_j = \frac{\tau(\lambda_0,m,h)}{\tau(\lambda_j,m,h)}, \tag{2}$$

i.e.,

$$T_j = \frac{\lambda_0^{\,b}}{\lambda_j^{\,b}} \left[\frac{\displaystyle\int_{x_1}^{x_2} x^4 Q_{\text{ext}}(x,m) \exp[-b\lambda_0 x/2\pi] dx}{\displaystyle\int_{x_1}^{x_2} x^4 Q_{\text{ext}}(x,m) \exp[-b\lambda_j x/2\pi] dx} \right]. \tag{3}$$

The limits of each integration are $x_1 = (2\pi/\lambda)r_1$, $x_2 = (2\pi/\lambda)r_2$ and depend on the appropriate wavelength. The T_j $(j=1, \ldots, J)$ can be evaluated for various representative values of b and $m \equiv n - ik$, with $1.34 \leqslant n \leqslant 1.54$, $0 \leqslant k \leqslant 0.1$ and $8 \leqslant b \leqslant 30$.

The nonlinear dependence of T_j $(\lambda_0,\lambda_j,b,m,\ldots)$ on λ, b, n and k shown in Eq. (3) and illustrated in Figs. 1 and 2 permits, in principle, the reconstruction of the values of the parameters b, n and k from a set of three ratios of the optical depths $\tilde{T}_j$ measured at an appropriate set of wavelengths (λ_0,λ_j).

To illustrate this argument by a simulated numerical experiment, let us assume that $b=15$ and $m=1.34-i0.1$ and compute the optical depths at $\lambda_0=0.7$, $\lambda_1=0.575$, $\lambda_2=0.525$ and $\lambda_3=0.45\ \mu$m. These optical depths yield the ratios $\tilde{T}_1$, $\tilde{T}_2$ and $\tilde{T}_3$. We would now attempt to reconstruct the value of b and m from these three simulated measurements $\tilde{T}_j$.

We applied a simple numerical search technique to find the combination (b,m) which when substituted into (3) gives

$$T_j(\lambda_0,\lambda_j)=\tilde{T}_j(\lambda_0,\lambda_j),\ j=1,\ 2,\ 3, \tag{4}$$

where T_j are computed according to Eq. (3) for a given set (b,m), and $\tilde{T}_j$ are the simulated measurements. The results of the search method showed that only the combination $b=15$ and $m=1.34-i0.1$ satisfied Eq. (4). Similar results were obtained when the numerical experiment was repeated for the reconstruction of other values of b and m.

The use of a numerical grid search technique to determine (b,m) is advantageous in the analysis of this problem because it illustrates the uniqueness of the results unambiguously and in a way independent of the special properties of any mathematical inversion procedure. However, once the uniqueness of the solution is established other suitable nonlinear inversion techniques can be developed and applied.

The numerical search technique can be easily applied to recover (b,m) from a larger set of measurements $\tilde{T}_j$ with $J>3$. While this approach leads to redundant information about (b,m) in the absence of noise, it can be very useful, however, in the presence of relatively large noise in the data $\tilde{T}_j$ as we will see in the following section.

3. Error analysis

To study the stability and accuracy of the results in the presence of noise in the data and to study the advantages of using a larger number of measurements with $3\leqslant J\leqslant 9$, we performed the following numerical experiment: we assumed $b=15$ and $m=1.44-i0.03$ and computed the corresponding ratios $\tilde{T}_j\,(\lambda_0,\lambda_j)$ according to (3) for $\lambda_0=0.4$ µm and for nine values of λ_j in the range $0.4<\lambda_j\leqslant 0.7$ µm. Different levels of noise were added to $\tilde{T}_j$ and the accuracy of the reconstructed values of (b,m) was analyzed for $J=3,\ 6,\ 9$.

To set up the grid for the search technique, Eq. (3) was calculated for the discrete points $b=8(1)30$, $n=1.34(0.01)1.54$, $k=0(0.01)0.1$, and stored in the computer. The synthetic data were matched against these stored values. For each data point n and k ranged over the discrete set of grid points defined above and an interpolation scheme was used to determine the corresponding b. Thus, for each grid point with a fixed n and k, the J ratios T_j give rise to J interpolated values of b. The program calculates the variance of the set of J ratios of b to pick out the grid point corresponding to the minimum in the variance according to Eq. (4).

The results of our studies are as follows:

1) If the ratios $\tilde{T}_j$ were determined to five significant figures the errors in the inferred values of the parameters were $\Delta b<0.5\%$, $\Delta n\approx 0.01$ and $\Delta k\approx 0.005$, for $J=3$.

2) If the ratios were determined to four significant figures, the errors in the inferred values of the parameters were $\Delta b<0.5\%$, $\Delta n=0.01$ and $\Delta k=0.005$, for $J=6$.

3) If the ratios were determined to three significant figures the errors were $\Delta b=1\%$, $\Delta n=0.04$ and $\Delta k=0.02$, for $6\leqslant J\leqslant 9$.

4) When the ratios are determined to two significant figures $\Delta b=10\%$, $\Delta n=0.15$ and $\Delta k=0.04$, for $J=9$.

Further theoretical considerations will perhaps enable the tolerance of larger errors in the determination of the ratios. One possibility is to consider the grid defined by b and n and an interpolation scheme devised for k. Alternately, the grid defined by b and k can be used to interpolate for n. Consistency then demands that the same value of (b,n,k) be obtained from the three interpolations. While increasing the number of ratios will yield more information the assumption of the constancy of the index of refraction with wavelength may hold only over a narrow region (0.4 to 0.7 µm).

Nonlinear inversion programs should be investigated. Such techniques may tolerate larger errors in the measurements.

4. Conclusions

The results obtained here are independent of the vertical profile. In satellite occultation experiments this will therefore eliminate the tedious and difficult considerations due to the sphericity of the medium.

Yamamoto and Tanaka (1972, 1974), Bullrich *et al.* (1974) and Kondratyev (1973) have pointed out the critical role that the imaginary part of the refractive index plays in the estimates of global heating or cooling. Our analysis shows that the multi-spectral radiometer determines the imaginary part of the index of refraction with great precision.

The measurements that we require are ratios of two transmission measurements and, therefore, most of the systematic errors cancel out. While four significant figure accuracy is desirable in the ratios of optical depths, technology considerations indicate that a determination to three significant figures is attainable for multi-spectral measurements (at least 6 but preferably 9) in the absorption-free regions so as to determine the relevant optical parameters.

In the forthcoming GATE project a polarimeter and a multi-spectral radiometer will be based on the S. S. *Oceanographer*, to monitor the aerosol content of the tropical air masses. It is hoped that these two methods will yield results comparable to one another.

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