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OPTICAL CHARACTERIZATION OF
TROPOSPHERIC AEROSOLS

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RESEARCH DIRECTORATE

September 1987

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PREFACE

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OPTICAL CHARACTERIZATION OF TROPOSPHERIC AEROSOLS

1. INTRODUCTION AND BACKGROUND

With the increasing availability over the past two decades of high-speed electronic computers, complex calculations of optical extinction and scattering of electromagnetic radiation by atmospheric aerosols using the Mie theory¹ have become commonplace. The technology has advanced so swiftly that specialists measuring cloud extinction of aerosols often are unaware of advances being made simultaneously by other specialists measuring angular scattering patterns of aerosols with nephelometers. Such measurements and calculations can be done in numerous wavelength regions, but the technology which concentrated first upon the visible wavelengths now is expanding rapidly, especially into the regions of relatively high atmospheric transparency called 'windows' in the infrared. This technological expansion can be measured by a sampling of papers appearing in journals during the past year, discussing aerosol measurements or calculations of extinction,^{2,3,4,5,6,7,8,9} angular light scattering or nephelometry,^{5,10,11,12,13,14,15} lidar^{16,17,18,19} and related aerosol technology including thermal emissivity²⁰ and calibration of optical particle counters.²¹

Several trends have been noted in all of this work. First, the utility and information content of simple extinction measurements has tended to be overlooked in the rush to exploit scattering techniques. Second, the polarization techniques do not seem to include applicable techniques, some new, that can be applied both to extinction and to polarization measurements. Third, if taken in combination at this point in time, the overall technology might be further advanced than is realized by workers in the individual disciplines. For example, remote characterization or quantitative analysis of tropospheric aerosols seems a distinct possibility, both by conventional transmission (extinction) spectroscopy and by angular scattering or back-scattering techniques. This report examines these possibilities by first reviewing the technology of aerosol extinction spectroscopy.

The Beer-Lambert equation for atmospheric aerosols can be written:

$$-2.3 \log(T_\lambda) = a_\lambda CL \quad (1)$$

where for a given wavelength λ (μm), T_λ is the fractional transmittance, a_λ is the optical mass extinction coefficient of the aerosol (m^2g^{-1}), C is the aerosol mass concentration (gm^{-3}) and

L is the optical path length (m). Some form of Equation 1 has been used by spectroscopists virtually since the birth of spectroscopy and the equation has become so well known that the power of simple extinction measurements of aerosols often is not fully appreciated. For example, using suitable multi-wavelength techniques, extinction measurements can contain sufficient information to allow direct determination of the mean size of a spherical droplet aerosol distribution or even remotely to characterize chemical reactions in cloud droplets at isosbestic wavelengths, without the use of polarized radiation or other more sophisticated techniques.^{22,23,24,25}

It is not surprising, however, that polarization techniques have received heavy emphasis since their exploitation in the early 1940's to characterize military fog-oil smoke mean droplet diameter based, in part, in earlier work at the U.S. Bureau of Standards and by Stratton and Houghton using the Mie theory.^{26,1} The polarization ratio then was measured only for transparent spheres for which, if the complex index of refraction m_λ is given by $(n - ik)_\lambda$ where n_λ is the real index and k_λ is the imaginary index (closely related to the absorption coefficient), then $k_\lambda = 0$. Polarization measurements can be used to determine particle size distribution in addition to mean size of aerosol droplet distributions. This was recognized and the technique was developed in the 1960's.^{27,28,29} By 1969, the techniques of particle size distribution measurements had been extended, as discussed in Kerker's classical book on light scattering. This book gives an excellent review of the technology as it existed then.³⁰

In recent years, there is an increased awareness that aerosol particle absorption, e.g., in the infrared where $k \neq 0$, can be interpreted in new ways utilizing not only extinction measurements or polarization measurements, but the two together.^{22,23,24,30} The author concurs that this is a promising field for new research and this Laboratory is developing an expanded capability for scattering and polarization measurements to complement ongoing research in extinction spectroscopy of aerosols.

2. AEROSOL EXTINCTION MEASUREMENTS

It is known that the variation of k_λ from values near zero in the visible wavelengths to significant ones in the infrared wavelengths provides a method for the determination of aerosol mean droplet size and mass concentration from simple extinction measurements if two or more wavelengths are selected properly for observation. For example, at the He:Ne wavelength $\lambda = 0.63 \mu\text{m}$, $k_\lambda \sim 0$ and typical water fog droplets have diameters D_μ (in μm) such that $D_\mu \gg \lambda$ or, to use Kerker's criterion, the size parameter $\pi D_\mu / \lambda \geq 2.0$ so that the conditions for geometric scattering exist. This leads to a constancy of the product $\alpha_\lambda \cdot D_\mu \rightarrow 3.0-3.2$, where α_λ is defined in Equation 1. At the same time,

the product $\alpha_\lambda \cdot D_\mu$ decidedly is not constant with D_μ at wavelengths where $k \neq 0$ in the infrared. This is illustrated in Figure 1, calculated from the Mie theory. In Figure 1, curves are shown for the infrared wavelengths of 8.5, 10.5 and 12.57 (or 4π) μm , as well as for a composite of wavelengths in the 8.5 - 12.57 μm region where extinction can be measured by a simple broadband transmissometer operating at 8-13 μm in the atmospheric window region. While operation at a specific wavelength such as the 10.6 μm CO₂ laser line for comparison to a visible wavelength such as the He:Ne line at $\lambda = 0.63 \mu\text{m}$ has advantages, a broadband infrared reference band also can be used. In fact, related findings, using a filtered light source in the visible at $\lambda = 0.515 \mu\text{m}$ and a 9-12 μm broadband transmissometer to study the formation and dissipation of water fogs, were reported at least as early as 1970.³¹

When the techniques of Carlon et al.,^{22,24} are extended to use the Mie theory to calculate functions of the transmittances T_λ at $\lambda = 0.63 \mu\text{m}$ and the broadband or composite wavelength band 8.5-12.57 μm versus water droplet diameter D_μ , curves like those shown in Figure 2 are obtained. It is seen that the calculated curves are widely separated. Thus, a sensitive method is provided for the determination of mean droplet diameter. Experimental points for a steam-generated water fog are shown in Figure 2. These give excellent agreement with calculated curves, for example $D_\mu = 8 \mu\text{m}$. This result recently was cited by Weinman et al.,³² as approximating Deirmendjian's³³ model C-1 (or vice versa), where D_μ is taken as the mass median diameter. These results seem credible. What is uncertain is the kind of distribution to which this diameter is most applicable. Kerker³⁰ has given an informative discussion of particle size distributions, and of transmission (extinction) and turbidity measurements. In this report, turbidity can be taken as the product $\alpha_\lambda \cdot C$ in Equation 1, having the units m^{-1} .

Justification for the use of composite of wavelengths (Figure 1 and abscissa, Figure 2) can be found by examining calculated curves and steam generated fog data points for the individual wavelengths $\lambda = 8.5, 10.5$ and 12.57 (or 4π) μm and plotting curves like Figure 2 as is done in Figure 3. Viewing the plots from top to bottom in Figure 3, it can be seen that the fog data gave best agreement with calculation for a mass median diameter (MMD) of 6 μm at $\lambda = 8.5 \mu\text{m}$, MMD = 10 μm at $\lambda = 10.5 \mu\text{m}$ and MMD = 6 μm at $\lambda = 4 \mu\text{m}$. In his discussions of particle size distribution functions and transmission measurements, Kerker³⁰ cautions that mean particle diameters obtained using methods like those discussed in the present paper may be adequate for most purposes if the distribution is sufficiently narrow, but strictly are valid only for monodisperse systems. By using a composite of wavelengths to obtain a mean diameter, one is averaging both over wavelength intervals and over particle size distributions. The results indicate that the technique works very well, especially under dynamic droplet growth or evaporation conditions where what

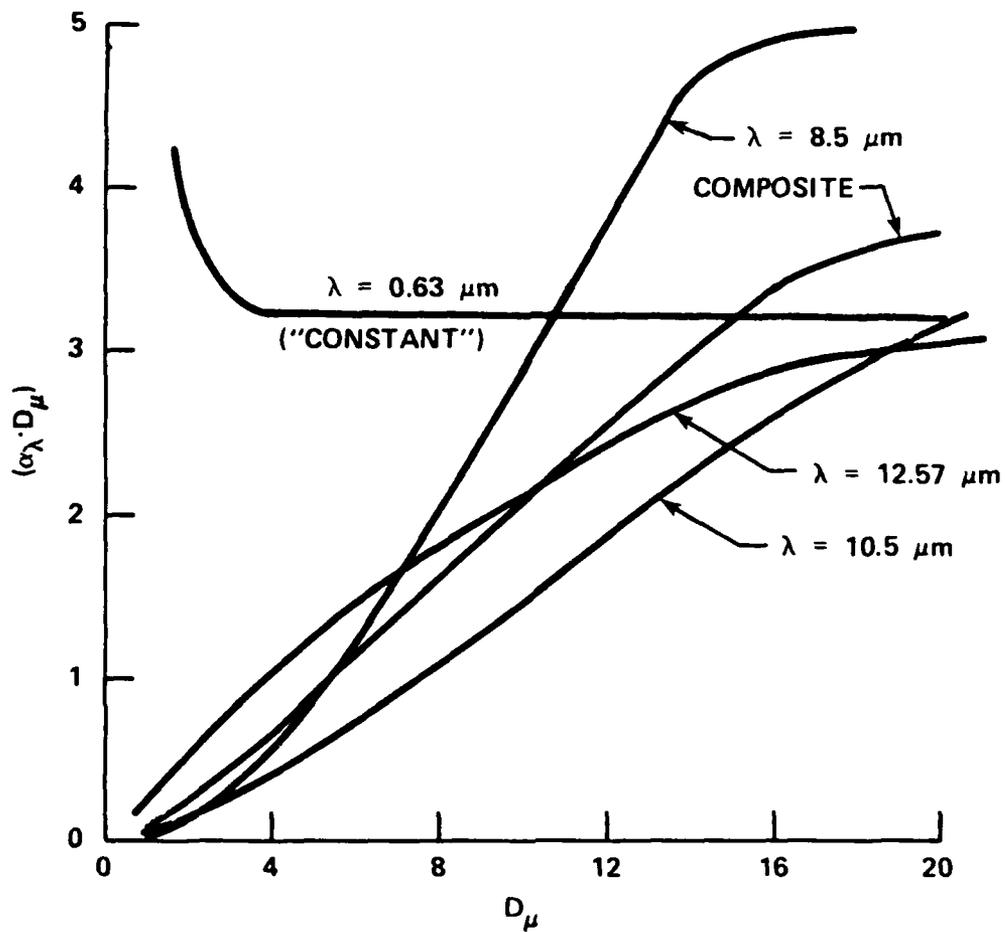


Figure 1. Values of $\alpha\lambda D\mu$ vs. $D\mu$ Calculated from the Mie Program for Water Using the Data of Hale and Query.³⁸ The Average Value of the 'Constant' Curve Is Shown; Actually the Function Is a Damped Oscillation.

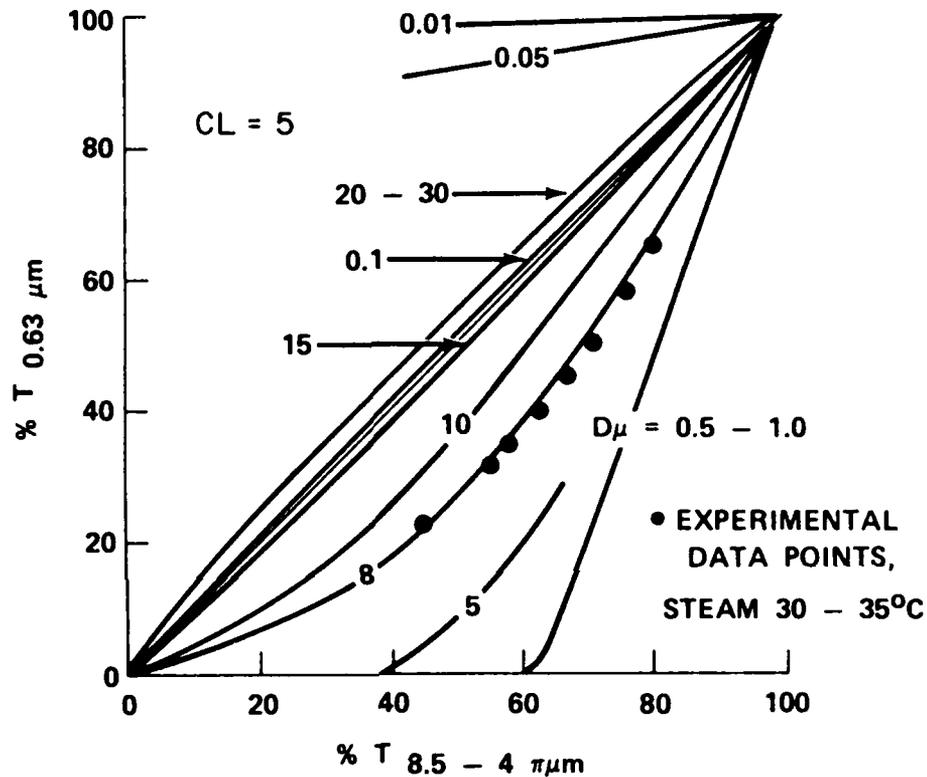


Figure 2. Comparison of Computed Curves Relating Visible to Infrared Optical Transmittance of Water Fog to Experimental Data Points for Cooling, Steam-Generated Water Fog Clouds; Note that Data Points Cluster Along the Curve $D\mu = 8 \mu\text{m}$ in Good Agreement with Gravimetric Data from the Same Experiment.

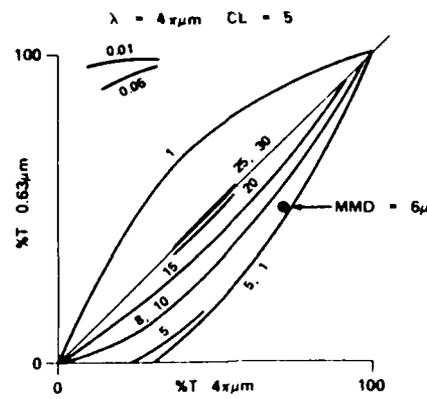
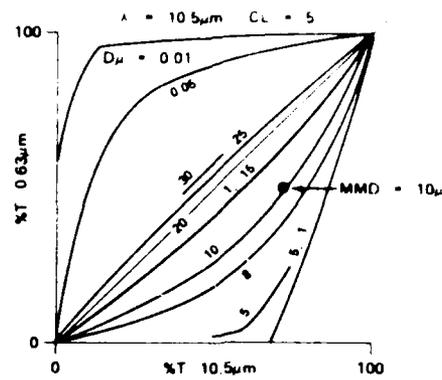
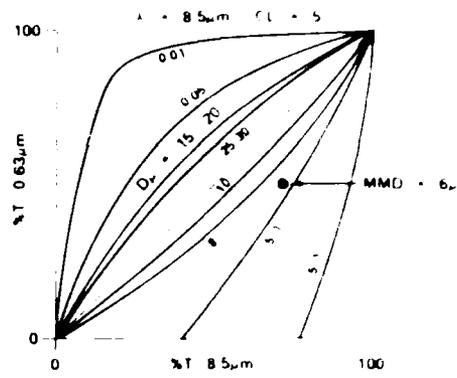


Figure 3. Curves for Three Infrared Wavelengths Comprising the Broadband Composite of Wavelengths Shown as the Abscissa of Figure 2.

is wanted is a real-time indication of approximate droplet size, e.g., in studies of developing or dissipating water fogs.^{31,34}

The highly variable dependence of α_λ upon λ is due to variations in $(n - ik)_\lambda$ in addition to which α_λ varies with $D\mu$ as was discussed previously. This is illustrated in Mie calculations for the extinction coefficient of water droplets (α_λ , ordinate) in Figure 4 vs. $D\mu$ for several wavelengths. At certain wavelengths, e.g., $\lambda = 12.5 \mu\text{m}$, the combined contributions of refractive (η_λ) and absorptive (k_λ) components of water droplet extinction lead to values of α_λ that virtually are independent of droplet size. Experiments seem to confirm that a $12.5 \mu\text{m}$ is nearly constant with $D\mu$, suggesting that the liquid water content C in Equation 1 of a tropospheric optical path could be monitored by a simple $12.5 \mu\text{m}$ transmissometer with good precision for droplet sizes of up to about $15 \mu\text{m}$.^{24,35} In this application, however, care must be taken to account for absorption due to hydrogen bonding in molecular clusters (water clusters) in the vapor leading to absorption easily confused with droplet absorption.²

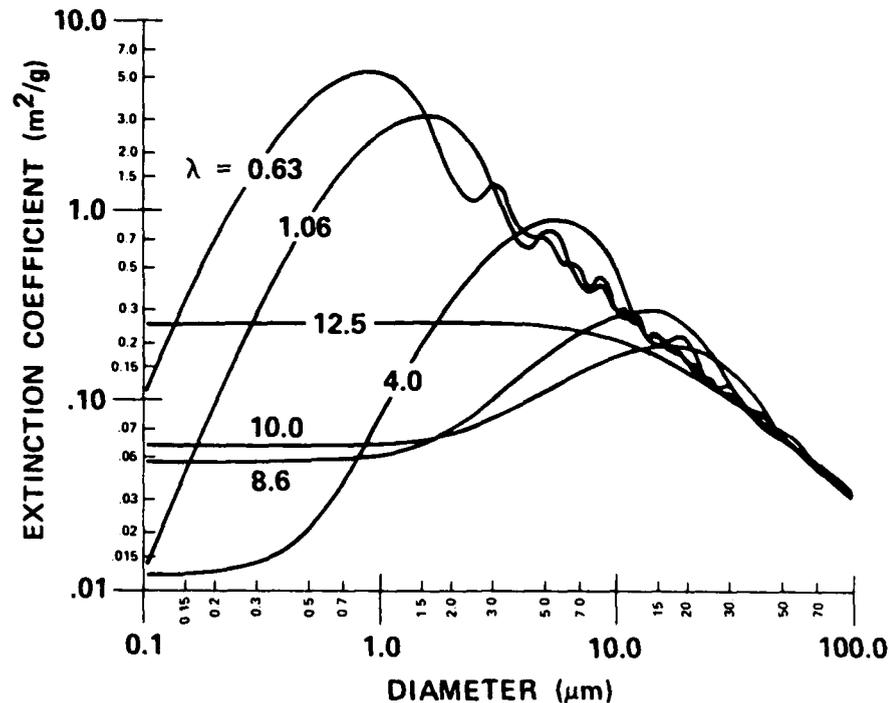


Figure 4. Calculated Curves from the Data of Hale and Querry³⁸ for Water Droplets Showing α_λ vs. $D\mu$; Note Constancy of $\alpha_{12.5}$ at All Diameters Up to $D\mu \sim 15 \mu\text{m}$.

At wavelengths called 'isosbestic points',²⁵ the complex indices $(n - ik)_\lambda$ of some liquids are such that α_λ remains constant even though the liquid solutions comprising the droplet aerosol vary widely in their chemical composition, e.g., for droplets of orthophosphoric and related acids that might be formed by burning phosphorus in air of varying relative humidity such that the solute (acid) concentrations vary from near zero to near 85% by weight. In Figure 5, $\lambda = 11.4 \mu\text{m}$ is an isosbestic point calculated from refractive index measurements of pure orthophosphoric acid (H_3PO_4) in water.^{36,37} Real atmospheric smokes, obtained by burning phosphorus, may contain mixtures of acids depending on the rates of combustion in the presence of varying humidities, and so may not give experimental spectra like those for pure H_3PO_4 in Figure 5, where $\alpha_{11.4}$ remains essentially constant at $0.13\text{--}0.15 \text{ m}^2\text{g}^{-1}$, while acid concentration varies from near zero to 85% by weight. This will be investigated. Because of the constancy of $\alpha_{11.4}$ compared to the wide range of values of, say $\alpha_{9.7}$ (see Figure 5) with solute concentration, remote characterization of the aerosol becomes a possibility if something is known about the droplet constituents. It can be shown that:

$$\alpha_\lambda \approx 4\pi k_\lambda f(m_\lambda) / \lambda \rho \quad (2)$$

where ρ (g cm^{-3}) is the droplet solution (acid) density i.e., mass density, and:

$$f(m_\lambda) = \left[\frac{9n}{(n^2 + k^2)^2 + 4(n^2 - k^2) + 4} \right]_\lambda \quad (3)$$

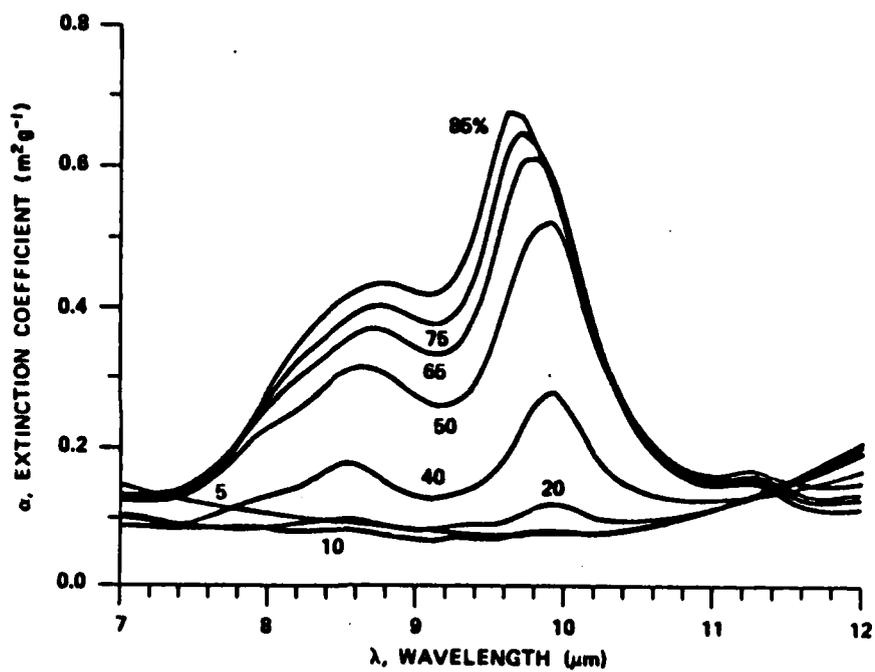


Figure 5. Computed Values of α_λ for Liquid Droplet Aerosols Comprising Several Concentrations of Orthophosphoric Acid (H_3PO_4) in Water, from Optical Constants of Querry and Tyler.³⁷

where $m_\lambda = (n - ik)_\lambda$ and the function of Equation 3 is plotted in Figure 6 for values of η_λ marked on the curves and ranging from 0.1 to 4.0, and for values for k_λ shown on the abscissa. It is emphasized that the above discussion applies to Rayleigh-scattering smokes where $D_\mu \ll \lambda$ as clearly is the case for orthophosphoric or other acid aerosols (smokes) produced by burning phosphorus in moist air.

Often it is useful to plot extinction coefficients or fractional transmittances versus D_μ rather than versus the size parameter $\pi D_\mu / \lambda$, for specific wavelengths. Two such calculated plots are shown for water droplets in Figure 7 and 8.³⁸ In Figure 7, the curves are calculated for a product $C \times L = 5.0$ (Equation 1) and for wavelengths, reading from top to bottom on the left-hand ends of the curves, of $\lambda = 0.63, 8.5, 10.5$ and $4 \pi \mu\text{m}$ (the same wavelengths considered in Figures 1 - 3). In Figure 8, the curves are calculated for $\lambda = 10 \mu\text{m}$ to show the functions vs. D_μ of total extinction, and of the scattered and absorbed components that are summed to give total extinction, i.e.,

$$\alpha_T = \alpha_S + \alpha_A \quad (4)$$

where the subscripts indicate total (T), scattering (S) and absorptive (A) extinction.

Sassen²³ recognizes that aerosol cloud extinction measured in conjunction with angular scattering measurements could lead to remote sensing of cloud composition by using two wavelengths like $\lambda = 0.63$ and $10.6 \mu\text{m}$.²³ But the present discussion indicates that such remote characterization could solely result from cloud extinction measurements at two closely-spaced infrared wavelengths, one of them an isosbestic point. Additional data might be obtained by combining extinction and angular scattering measurements at an isosbestic wavelength. Optical parameters can carry only so much information, but the idea is provocative enough to warrant an investigation.

Other investigations for remote characterization of tropospheric aerosol clouds are possible, such as those based on effects peculiar to Christiansen wavelengths.³⁹ A complete discussion is given by Carlon where the latter reference discusses aerosols ranging in size from molecular clusters to water cloud droplets.^{2,39}

3. POLARIZATION AND ANGULAR SCATTERING MEASUREMENTS

Kerker³⁰ has given an excellent review of techniques to determine particle size distributions using these measurements and I will not elaborate except to raise a few pertinent points. Sassen²³ has verified that angular scattering patterns calculated from the Mie theory can be verified rather closely by experiment not only for spherical particles but for irregularly-shaped ones as well. Thus polarization or angular scattering techniques

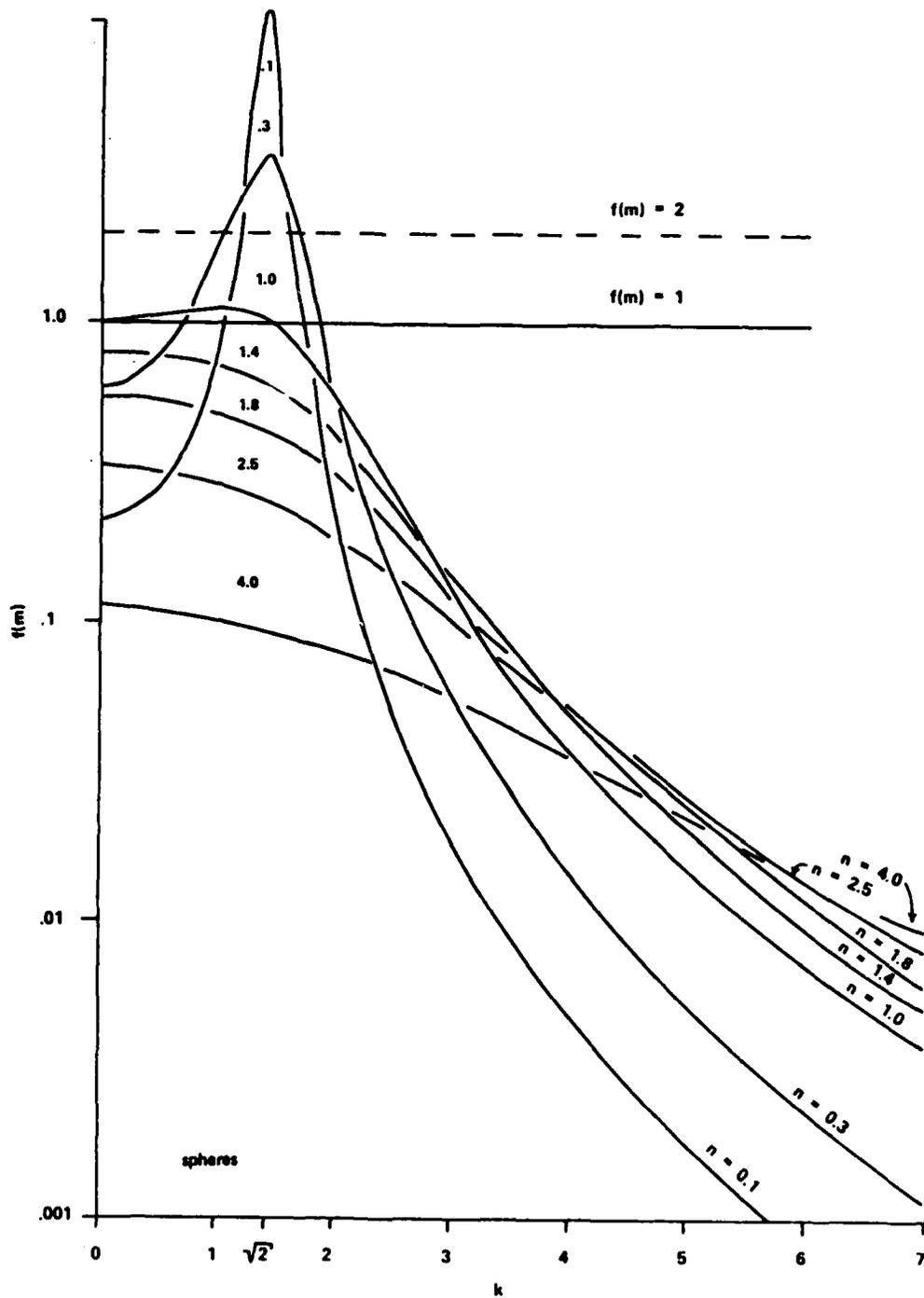


Figure 6. Plot of Equation (3) for η_λ Values of 0.1-4.0 Shown on the Curves and k_λ Values Shown on the Abscissa; for Rayleigh-Scattering Aerosols 'Only' ($D_\mu \ll \lambda$).

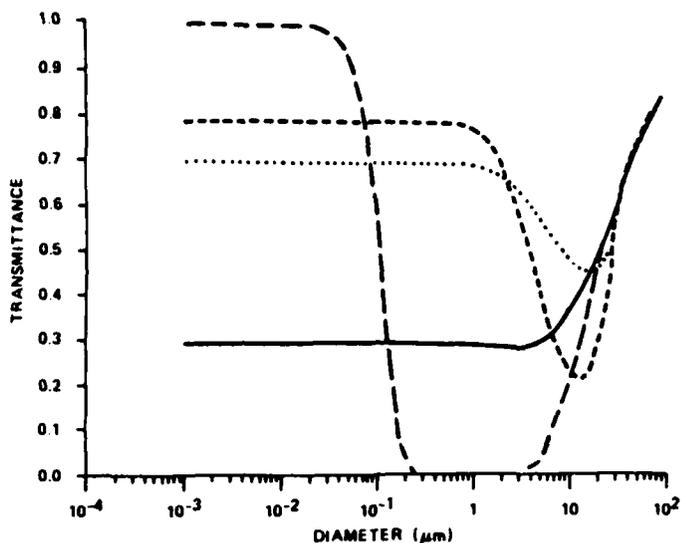


Figure 7. Fractional Transmittance for a $C \times L$ Product = 5.0 (Equation 1) of Water Fog vs. Droplet Diameter for Wavelengths, Reading from Top to Bottom of the Left-Hand Ends of the Curves, of $\lambda = 0.63, 8.5, 10.5,$ and $4\pi \mu\text{m}$, which Are the Wavelengths also Considered in Figures 1 - 3.

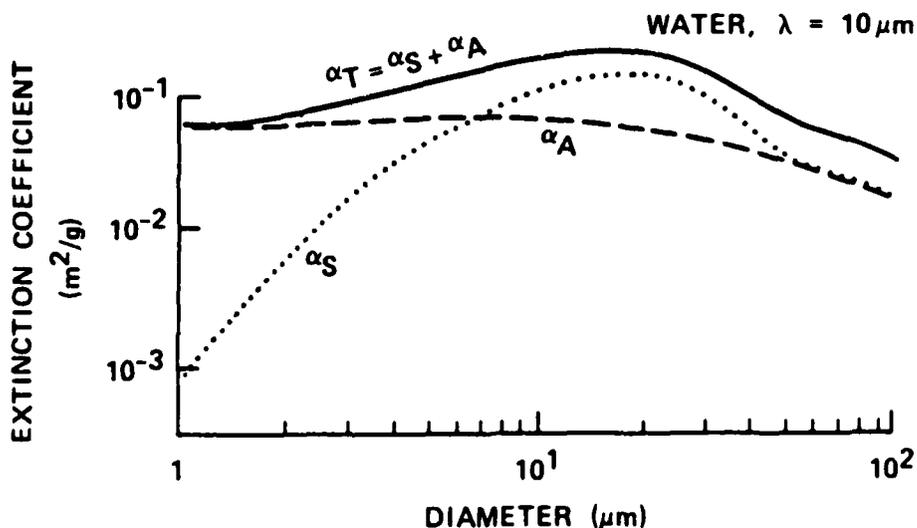


Figure 8. α_λ vs. $D\mu$ Calculated from Mie Program Using Data of Hale and Querry³⁸ at $\lambda = 10 \mu\text{m}$ for Water Droplets; the Subscripts T, S, and A Refer to Equation (4) and Definitions.

combined with some of the recent advances in extinction measurement technology discussed here should provide new combinations for the remote characterization of tropospheric aerosols.

The polarization method for evaluating the distribution of sphere sizes, utilizes polarization of scattered radiation obtained from a monochromatic light source at various angles of observation.³⁰ The intensity of the component of the scattered light, whose electric vector vibrates perpendicular and parallel to the plane of observation, $I_1(\theta)$ and $I_2(\theta)$, is measured at a number of angles, where θ is the scattering angle measured from the direction of the incident light beam. A polarization ratio, $P(\theta)$, is defined such that:

$$P(\theta) = I_2(\theta) / I_1(\theta) \quad (5)$$

For example, $P(\theta)$ can be calculated as a function of for any assumed particle distribution such as the zeroth order logarithmic distribution (ZOLD) and for any standard deviation, O_0 , ranging from $O_0 = 0$ (i.e., a monodisperse aerosol), to values of 0.3 or more where the curves flatten and the method approaches its limit of usefulness. Figure 9 shows the family of curves calculated for the He:Cd wavelength $\lambda = 0.4416 \mu\text{m}$, $D_\mu = 0.3 \mu\text{m}$, $\eta_\lambda = 1.484$ and $k_\lambda = 0$ so that $m_\lambda = (1.484 - i \cdot 0) \lambda$ and the density is $\rho = 0.98 \text{ g cm}^{-3}$. The curves are for dioctyl phthalate (DOP), a liquid commonly used for test aerosols.² Agreement between calculation and measurement using the polarization method is good to excellent, often within a few percent.

The effect of the wavelength of the monochromatic light upon the function shown in Figure 9 is of special interest. This has been investigated^{27,28} for visible and near infrared wavelength, including cases such as vanadium pentoxide (V_2O_5) for which strong absorption occurs at shorter visible wavelengths, i.e., $k_\lambda \neq 0$.²⁹ Special techniques using extinction measurements in the infrared where $k_\lambda \neq 0$, suggest that combinations of visible and infrared wavelengths can optimize desired parameters using polarization techniques. Since the size parameter $\pi D_\mu / \lambda$ depends on wavelength, a change in the wavelength of the incident light will cause a shift in the position of the curve peaks in Figure 9, larger values of λ shift the peaks to the right. Multi-wavelength observations have been used to show that particle size distributions obtained at different wavelengths are consistent, but when the wavelength spacing is as great as $\lambda = 0.63 \mu\text{m}$ and $10.5 \mu\text{m}$, additional work is needed to confirm this and also to determine whether the useful range of O_0 can be extended beyond 0.3 in measurements using the polarization method.³⁰

The scattering ratio method for the determination of particle size distribution is a variation where the angle of observation is held constant and the polarization ratio is

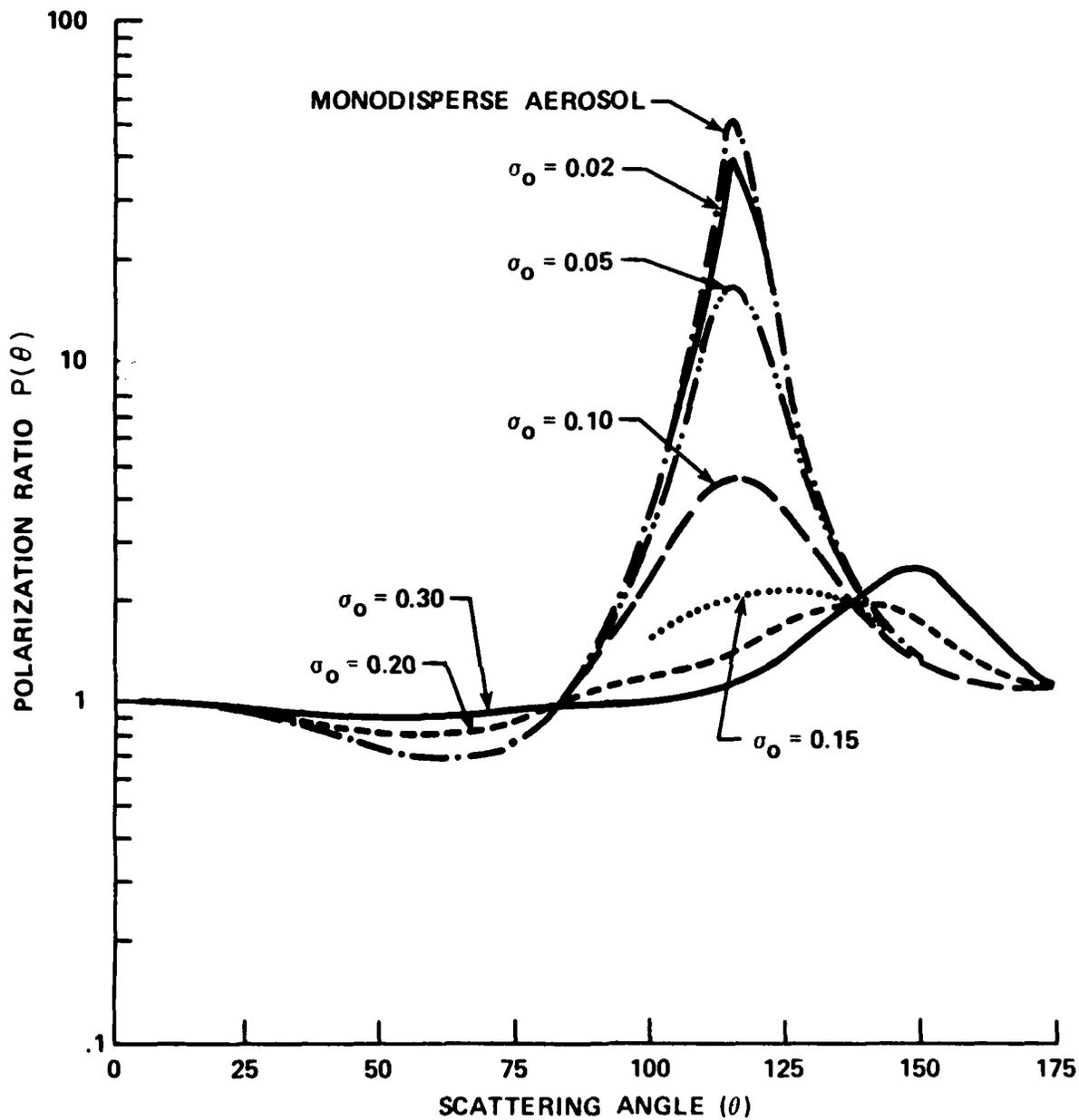


Figure 9. Polarization Ratio $P(\theta)$ vs. Scattering Angle θ for DOP.

measured or calculated over a spectrum or over a number of discrete wavelengths.^{40,41,42} The method was perfected by using monodisperse, polystyrene, latex spheres and known combinations of single sphere sizes to give mixtures of known size distributions checked by electron microscopy. The scattering ratio method gives an alternative way of studying the variation of O_0 at some D_μ with λ at some constant θ . The same thing can be done by using the polarization method directly and plotting, e.g., Figure 9 with λ as the abscissa at constant θ .

Turbidimetric particle size distribution methods invariably are similar to those of Wallach et al.⁴⁰ Very recently Nelson⁴³ proposed a method by which calculated scattering cross sections from the Mie theory are compared to experimental extinction data using monodispersed, polystyrene, latex spheres in distilled water as a calibration means. A computer program is used to null the error versus wavelength, forcing a unique solution for the size distribution which is taken as bimodal to account for coagulation. The method also is said to yield particle volume concentration and the scattering cross section function, $(Q)_\lambda$. Extrapolation of the technique to tropospheric aerosols remains to be demonstrated.

4. SUMMARY AND CONCLUSIONS

This report has concentrated on advances that have been made in the remote characterization of tropospheric aerosols by extinction and turbidimetric methods over the decade that has elapsed since the publication of Kerker's classic text.³⁰ The technology has been updated, including results not previously published, with improved techniques in polarization and angular scattering methods. Perhaps most importantly, the author has tried to support the argument that if all methods presently available in this technology are taken into account at this time, our ability to remotely characterize or quantitatively analyze atmospheric aerosols is further advanced than workers in the individual light measurement disciplines realize.

It is concluded that by combinations of well established and recently developed light scattering techniques, investigators can undertake promising new areas of research. Some examples include combinations of extinction and polarization measurements with computer programs to study isosbestic or Christiansen wavelengths, and optimization of desired parameters as functions of wavelength or particle size distribution.

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