Complex Index of Refraction of Airborne Fly Ash Determined by Laser Radar and Collection of Particles at 13 km

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ABSTRACT

On 30 September 1970, the National Center for Atmospheric Research (NCAR) obtained data on the vertical distribution of particulate material over Boulder, Colo., from laser radar soundings and simultaneous airborne particle collections. A layer of particulate material at about 13 km was of special interest. Particles in this layer differed from normal tropospheric particles and were probably fly ash created by forest fires in California during the previous week. A technique for determining the complex index of refraction of atmospheric particles has been applied to the 13-km data. By assuming the real part of the refractive index to be 1.55, the imaginary part (the absorption parameter) is estimated to be 0.044 ± 0.011.

1. Introduction

Many mechanisms are responsible for the continuous injection of particulate material into the atmosphere. Natural processes contribute such material as volcanic dust, meteoritic dust, spores and seeds, and particles of sea salt, soil, and ash from forest fires. Man's activities contribute such materials as fly ash from smoke stacks, mineral particles from industrial sources, and dusts from mining or milling operations. With time, the particulate material may be considerably modified from its original form by coagulation, sedimentation, and other processes. Generally speaking, these processes tend to reduce the number of very large and very small particles, and to leave particles of intermediate size of approximately 0.1–1.0 μm radius. Particles in this size range produce significant optical effects; for detailed studies ranging from remote measurements of the amount of particulate material released by industrial sources, to calculations of radiative transfer in the global atmosphere, it is important to be able to evaluate these effects.

The fundamental contribution to general scattering theory has been a solution for scattering by homogeneous spherical particles of arbitrary size and composition (Mie, 1908). Scattering coefficients specified by Mie's solution are related to the size and composition of spherical particles through the size parameter $\alpha = 2\pi r/\lambda$ (r is the particle radius and λ the wavelength of the incident radiation) and the complex index of refraction, $m = n - in'$, in which the imaginary part is an absorption parameter. Comprehensive discussions of the application of Mie scattering theory to analyses of light scattered by small particles can be found in a variety of standard references (see, for example, van de Hulst, 1957).

Except for very specific aerosol populations, such as a mist of suspended liquid droplets, Mie scattering calculations should be used only with reservation in quantitative applications, since the particles are often known to have non-spherical shapes and to be of mixed composition. However, considerable insight can be obtained by applying Mie scattering calculations to systems of polydisperse homogeneous spheres having an assumed index of refraction and size distributions approximating those observed for atmospheric aerosols. Analyses based on these calculations can be utilized for interpreting aerosol scattering data in terms of an equivalent distribution of Mie scatterers.

Most quantitative analyses of atmospheric scattering phenomena suffer from the fact that even though the real part n of the index of refraction of the particles is reasonably well known, little information is available about the absorption parameter n'. This is by no means related to a lack of interest in the absorptive characteristics of the aerosol; rather it reflects the extreme difficulty associated with determination of the absorption of small particles when they are actually suspended in air. Eiden (1971) has discussed an entirely optical method of determining the complex index of refraction by analyzing the degree of polarization, the ellipticity, and the position of the ellipse described by the electrical field vector of the polarized portion of the light scattered by spherical particles. However, investigators using the technique described by Eiden may experience numerous experimental difficulties in

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obtaining an actual measurement. Some of these difficulties were pointed out by Eiden; others may be more subtle and may cause systematic errors which will adversely affect the interpretation of results based on measurement techniques in which the systematic error would not, in effect, cancel out. As an example, there is mounting evidence that existing measurements (typically made several decades ago) of the depolarization of light scattered by air molecules may incorporate substantial errors due to the inability of the experimenters to reduce the background level of atmospheric aerosols to the point where they had negligible effects on the measurements of molecular scattering (Shardanand, 1971).[2]

We have devised a scheme for determining the complex index of refraction for atmospheric aerosols. Using established measurement techniques—laser radar sounding and study of particles collected with airborne impactors—we have performed preliminary observations to demonstrate the feasibility of our scheme. The following sections deal with descriptions of our procedure and analysis techniques used in determining the imaginary part of the refractive index for a specific type of atmospheric aerosol for which the real part is assumed to be known.

2. Particle collection

Samples of atmospheric particulates were collected using the calibrated single-stage jet impactor described by Blifford and Ringer (1969), which was installed in the NCAR Sabreliner jet aircraft. The original intake system was modified for operation in the pressurized cabin so that ambient air entered through an isokinetic inlet into an auxiliary chamber within the cabin, and was directed to the impactor through a second isokinetic sampling nozzle placed in the laminar flow portion of the chamber. An effort was made to avoid discontinuities which could result in particle deposition

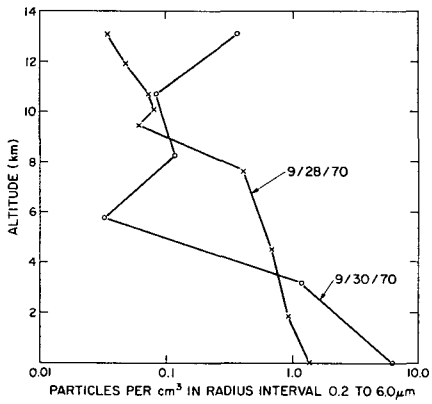

FIG. 1. Vertical profiles of number concentration for particles in radius interval 0.2–6 μm at Scotts Bluff, Nebr., on 28 September 1970, and Boulder, Colo. on 30 September 1970.

[2] Private communication.

TABLE 1. Total number of particles (cm^{-3}) in radius interval 0.2–6 μm at 13 km.

Date	Location	Particle concentration
28 September 1970	Scotts Bluff, Nebr.	0.03
30 September 1970	Boulder, Colo.	0.37
8 October 1970	Boulder, Colo.	0.42
30 October 1970	Scotts Bluff, Nebr.	0.26
7 December 1970	Scotts Bluff, Nebr.	0.09
11 January 1971	Scotts Bluff, Nebr.	0.40
10 March 1971	Scotts Bluff, Nebr.	0.44
	Average	0.29

in the intake system. The air flow rate of 12.5 liters min^{-1} was obtained by ram pressure at the inlet and by the use of a carbon vane positive displacement pump. The particles were collected on standard microscope slides, and the size and number distribution determined using the photomicrographic technique described by Blifford and Ringer.

Fig. 1 compares vertical profiles of number concentration for particles in the size interval 0.2–6 μm obtained at Scotts Bluff, Nebr., on 28 September 1970 with those of 30 September 1970 at Boulder, Colo.; the two locations are close enough that, for a given day, similar aerosol data should be observed at each location. On 28 September the particle concentration decreased nearly exponentially with altitude; on 30 September, the concentration at 13 km was greater by approximately a factor of 10. Table 1 indicates that the total concentration of 0.37 particles cm^{-3} was not exceptionally high on 30 September but was greater than the average of 0.29 particles cm^{-3} for all samples at 13 km.

Inspection of size distribution curves, however, shows that the increase in total number on 30 September was due almost entirely to an increase in the number of larger particles. The logarithmic distributive functions $dN/d(\log r)$ for aerosols collected at an altitude of 13 km over a period of four months at Scotts Bluff are shown in Fig. 2. The indicated errors include the sampling error based on the number of particles counted, and combined instrumental and measurement

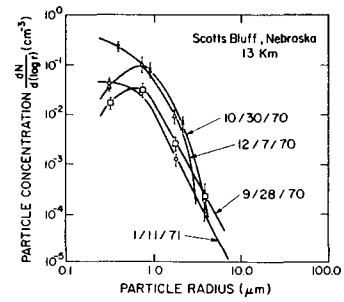

FIG. 2. Size distribution of particles collected at 13 km altitude during a 4-month period at Scotts Bluff, Nebr.

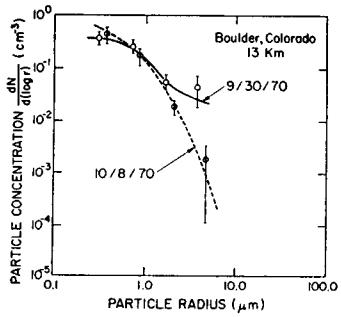

FIG. 3. Size distribution of particles collected at 13 km altitude at Boulder, Colo., on 30 September and 8 October 1970.

errors. In comparison with samples obtained at lower altitudes (Blifford, 1970), the main feature distinguishing the present data is the rapid decrease in particle number for particles larger than a few microns. Since a variety of atmospheric processes (sedimentation and rainout, for example) produce relatively rapid and efficient removal of particles in this size range, older aerosols such as would be present in the stratosphere are expected to have characteristic size distributions that are deficient in large particles. Fig. 3 shows the size distribution functions for particles collected at 13 km over Boulder on 30 September and 8 October 1970. It can be seen that the 30 September sample contained of the order of 100 times as many particles of about 5 μm radius as on the other occasions.

Examination of photomicrographs of the particles collected at 13 km on 30 September 1970 revealed a high concentration of combustion product residues similar to fly ash. We are reasonably certain that the extraordinary increase in the concentration of large particles was due to forest fires which were burning at that time in California although we have not confirmed the origin of the particles by determining their trajectories from analyses of weather charts. While their origin is intrinsically interesting, it is not germane to the discussion of our technique for evaluating the complex index of refraction of airborne particles.

3. Laser radar soundings

Vertical profiles of the backscattering cross section of the atmosphere were measured at Boulder during the evening of 30 September 1970. Data were obtained with the NCAR laser system described by Schuster (1970). During normal operation, the laser generates pulses of light of 20 nsec duration, having approximately 1–2 J of optical energy at a pulse repetition rate of about 4 min^{-1}. As the pulse propagates through the atmosphere, backscattered light is collected by a 60-inch (1.5 m) searchlight mirror modified to serve as a Cassegrain telescope. The energy collected by the receiver is collimated and then directed through a narrow band interference filter to decrease the intensity of background light from the sky. The intensity of light at the laser wavelength (0.6943 μm) was measured with an EMI 9558A photomultiplier cooled with dry ice. The current generated by the photomultiplier was fed into a gain-switching preamplifier and sent into a high-speed data acquisition system that sampled the photomultiplier signal at 660-nsec intervals and digitally recorded the signal on magnetic tape. The digitized signals were subsequently analyzed on the CDC 6600–7600 computer at NCAR.

For a dust-free atmosphere, the measured intensity of the light backscattered from the laser beam would exhibit a smooth, monotonic decrease with altitude and would provide data on the density of the atmosphere for each range interval sampled by the data acquisition system. Since particulate material suspended in the atmosphere tends to have particle sizes of approximately 0.1–1 μm radius, and since particles of this size produce significant optical effects for visible radiation, the measured backscattering cross section of the atmosphere would be increased for those altitude regions where backscattering from dust particles is significant relative to the backscattered signal from the molecular atmosphere.

The intensity of echoes for an optically thin layer of homogeneously distributed scatterers, expressed in terms of the expected rate of emission of photoelectrons per transmitted pulse, is

$$\frac{dn_p}{dt}=\frac{WK_1K_2A\eta\lambda}{8\pi h}\cdot\frac{K_a^2\Sigma}{R^2}, \tag{1}$$

where W is the transmitted energy per pulse, K_1 and K_2 the efficiencies of the receiving and transmitting systems, A the collecting area of the receiving telescope, η the quantum efficiency of the photocathode, λ the wavelength, K_a the atmospheric transmission, Σ the collective radar cross section of the scatterers per unit of volume, R the range, and h Planck's constant. The above "radar equation" is derived elsewhere (Grams, 1966); note that only three terms in the equation are range-dependent, namely, K_a, Σ and R. Σ is determined by the contributions to backscattering from the various atmospheric constituents. For a dust-free atmosphere, we would have a radar cross section

$$\Sigma_R=N\sigma_R,$$

where N is the molecular number density and σ_R the radar cross section of an individual air molecule, evaluated in accordance with Rayleigh scattering theory. Thus, the laser radar would be expected to measure a signal that is directly proportional to the atmospheric number density and the square of the atmospheric transmission, and inversely proportional to the square of the distance to the scattering volume. To interpret the measured laser radar signals, most investigators compute a ratio between the signal received and the signal expected from a dust-free

atmosphere, and use this ratio to locate the regions where dust scattering can be detected (Grams, 1970). Usually, the atmospheric number density is obtained from profiles tabulated in published standard atmospheres in order to maintain the logistic simplicity of the laser radar measurement technique. For this experiment, however, we were able to derive actual data of the density profile from measurements aboard the NCAR Sabreliner aircraft at each of the sampling altitudes. Laser radar data for time periods concurrent with the particle collection time intervals are presented in Fig. 4 along with values of the calculated scattering ratios at each of four sampling altitudes. The data are plotted in accordance with Mie scattering calculations for homogeneous spherical particles of refractive index 1.5 with particle size distributions matching those of the collected atmospheric particles. We have plotted the results of averaging scattering ratio profiles from all consecutive laser radar pulses obtained during each sampling interval. (The two calculated scattering ratios plotted with the bottom curve represent data taken during the first and second aircraft sampling intervals, since the laser system did not begin operating until approximately the beginning of the second sampling interval.) We obtained good agreement with calculated values at all altitudes except 13 km. The calculated value of the scattering ratio at 13 km for particles with refractive index 1.5 is significantly higher than the measured value. Apparently, dust at lower altitudes had an optical effect that agreed with that exhibited by the normal population of atmospheric aerosols, but that at 13 km did not. The particles at 13 km were probably fly ash and appeared to be significantly different from the normal particulate content of the atmosphere. In what follows, we use our measurements to determine the absorptive properties of the particles found at the 13-km level.

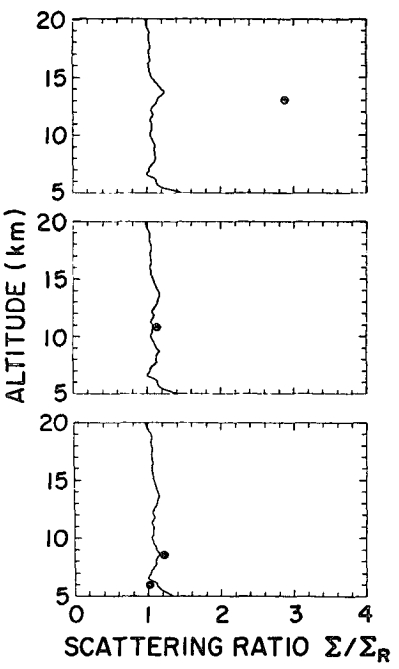

FIG. 4. Vertical profiles of scattering ratios at Boulder, Colo., on 30 September 1970. The points plotted with each curve are scattering ratios calculated by applying Mie scattering cross sections for refractive index 1.5 to the distributions of particle size observed at each altitude during concurrent time intervals.

4. Discussion

In order to evaluate the complex index of refraction, $m=n-in'$, for the particles observed at 13 km, we assume that the real part n is 1.55 in accordance with published values for fly ash (McCrone *et al.*, 1967). Values of the imaginary part n' are not available for fly ash, and we are therefore unable to calculate the expected signal for the observed distribution of particle sizes. We suggest, however, that our data can be used to affix a value to n' for the airborne fly ash. The approach is simply that of calculating the scattering ratio that would be observed for the observed particle distribution for varying values of the complex index of refraction, and selecting a value agreeing with the observed value. Fig. 5 shows the normalized radar cross section for homogeneous spherical particles with $n=1.55$ and with selected values of n'. In particular, we show the normalized cross section obtained for a case of no absorption, i.e., $n'=0$, and for cases in which $n'=0.005$, 0.01, 0.02, 0.05 and 0.1, bracketing the expected region for the imaginary part of the index. Note that, for the larger values of n', the backscattering cross section seems to approach a constant with increasing Mie size parameter. This constant, as might

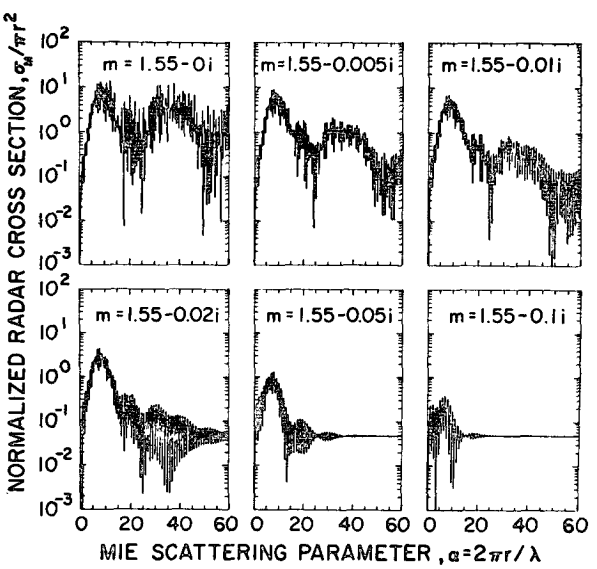

FIG. 5. Normalized radar cross sections for spherical particles having the indicated complex refractive indices, evaluated and plotted at Mie size parameter increments $\Delta\alpha=0.2$,

be expected, is the Fresnel reflection coefficient for a surface having the given complex refractive index.

Radar cross sections of the observed distributions of particle size were calculated from data on the number of particles in 48 logarithmically spaced radius intervals. An average radar cross section for each interval was obtained by calculating

$$\bar{\sigma}(\alpha_1,\alpha_2)=\frac{1}{\alpha_2-\alpha_1}\int_{\alpha_1}^{\alpha_2}\sigma(\alpha)d\alpha, \qquad (2)$$

with the definite integral evaluated in accordance with Simpson's rule. The values α_1 and α_2 are obtained by transforming the end points of each logarithmic radius interval to the corresponding values of the size parameter. Radar cross sections for the observed distributions of particle size were calculated by summing products of the number of particles observed in each radius interval times the average radar cross section of a single particle in that interval. To establish the maximum α increment that could be used to evaluate the average radar cross section for each radius interval, we calculated radar cross sections for the refractive index $m=1.5$ and the observed distributions of particle size with different integration increments to determine the point at which the accuracy relative to values calculated using the smallest interval ($\Delta\alpha=0.05$) began to deteriorate. In concurrence with the results of Dave (1969a, b), we found the maximum integration interval to be 0.2. We therefore used this increment to evaluate radar cross sections in subsequent calculations.

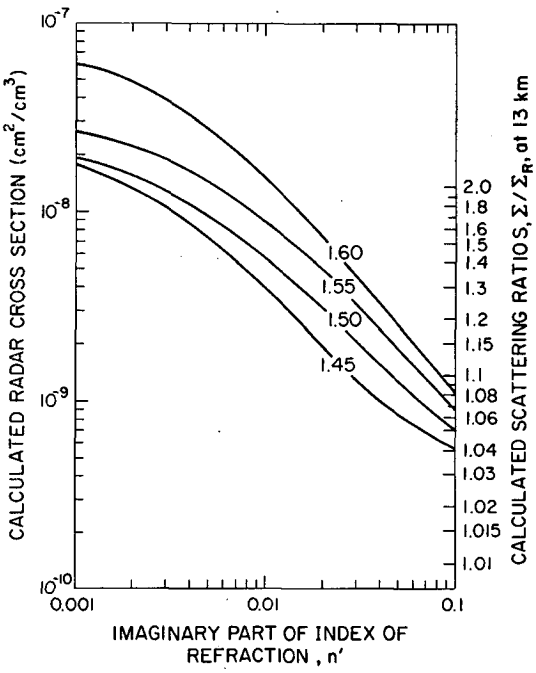

FIG. 6. Calculated radar cross sections at wavelength $\lambda=0.6943$ μm for the particle size distribution observed at 13 km at Boulder, Colo., on 30 September 1970 for varying values of the imaginary part of the refractive index. Numbers on the right refer to scattering ratios expected at 13 km altitude for the above conditions. Each curve refers to calculations for the indicated values of the real part of the refractive index.

Fig. 6 shows values of the total radar cross section for the distribution of particle sizes observed at 13 km for various sets of complex refractive indices having $n=1.45$, 1.50, 1.55 and 1.60. Each curve shows a smooth monotonic decrease with increasing absorption for the calculated values of the radar cross sections. A scattering ratio of 1.16 was measured at the 13-km level by the laser radar equipment. On the right side of Fig. 6, expected values of the scattering ratio, Σ/Σ_R, for a laser radar operating at 0.6943 μm, are plotted for particles having the calculated radar cross sections shown on the left side on the assumption that the molecular scattering is that expected from air having the 13-km number density. For $n=1.55$, the value of n' necessary to produce a scattering ratio of 1.16 is 0.044. An analysis of errors due to the expected statistical variability of the number of particles counted and the combined instrumental and measurement errors associated with both the particle sampler and the laser radar leads to an estimate of $\sim$40% error. By considering that the error results in a factor of 1.4 uncertainty, the error in the measurement of n' is 0.011. We therefore infer that, at least for use in interpreting laser radar data for this type of particle, the fly ash can be thought of as an ensemble of homogeneous spheres with refractive index $1.55-(0.044\pm0.011)i$.

The curves presented in Fig. 6 for varying values of the real part of the index of refraction can be used to determine the effect of inaccurately specifying n. Choosing $n=1.5$ or 1.6 leads, respectively, to refractive indices with $n'=0.030$ or 0.057; these deviations from the value of n' determined previously are of the same order as the measurement error. Since differences of the magnitude indicated by the above choices for n are not expected, the value of n' determined by this technique is relatively insensitive to small errors in the assumed value for the real part of the refractive index.

It is quite possible, although we have not yet performed experiments to determine the validity of this statement, that the method described above may also be applied to help interpret measurements taken by different types of instruments. Perhaps the largest concern that may be expressed regarding our scheme for determining the Mie absorption parameter is the fact that the particles collected at 13 km were of a very irregular shape whereas the Mie theory applies to spherical particles. Holland and Gagne (1970) show that the backscattering from irregular particles can deviate appreciably from that predicted by Mie theory. Although a departure from Mie theory would present a problem in applying the value of n' determined in this experiment to more general problems involving scattering at other angles, it would be consistent to use

values of n' derived by our scheme for interpreting other laser radar data.

5. Conclusions

The analysis presented here outlines a practical procedure for determining the characteristics of particles in the atmosphere which exhibit appreciable absorption. While we recognize that the imaginary part of the refractive index determined by our scheme may not necessarily be used for interpreting data obtained by different techniques, it certainly can be used to interpret other laser radar data.

Our research is currently being directed toward a much more complete study of the effect of aerosols on the propagation of electromagnetic radiation, and we expect in the future to be able to evaluate the extent to which aerosol absorption, quantitatively deduced from measurements of this type, can be applied to more general problems related to the propagation of electromagnetic radiation through the atmosphere.

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