Atmospheric Particulate Properties Inferred from Lidar and Solar Radiometer Observations Compared with Simultaneous In Situ Aircraft Measurements: A Case Study

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ABSTRACT

Particulate size and height distributions, complex refractive index and mass loading have been measured and inferred from direct aircraft and indirect lidar-solar radiometer observations made during a unique joint experiment conducted the week of 18 November 1974 in Tucson, Ariz. The aircraft and lidar-solar radiometer measurements were first analyzed independently and the results were then intercompared. Vertical profiles of particulate extinction obtained from the lidar (monostatic) and aircraft measurements were found to be in excellent agreement on both a relative and absolute basis. Lidar (bistatic and monostatic) inferences of particulate mass loading agreed favorably with the aircraft mass monitor measurements. The aircraft and lidar (bistatic) size distribution determinations were found to be similar in shape and agreed in absolute value within an order of magnitude. The mean particle refractive index inferred from the lidar (bistatic) measurements (n=1.40±0.000) agreed with the index of a significant fraction of the particles identified by electron microscope analysis of impactor samples collected with the aircraft.

1. Introduction

Since the first applications in the early 1960's of laser systems for atmospheric observations, a variety of theoretical and experimental techniques have been examined for improving the quantitative measurement capabilities of various lidar\(^1\) systems. Although substantial progress has been made in extracting useful information about atmospheric particulates from lidar signals, with very few exceptions (Grams et al., 1972; Grams et al., 1974) in situ measurements of particulate have not been quantitatively intercompared with remote lidar sounding observations.

An intercomparison experiment requires both sophisticated optical and airborne measurement systems—each of which requires for successful operation, the full attention of an active research group. The recent joint experiments reported by DeLuisi et al. (1976a, b) attest to the difficulty of quantitative aerosol-radiation (and lidar) intercomparison studies. This paper reports the results of a specific lidar-aircraft intercomparison experiment. The cooperative research was conducted as a subpart of existing studies at The University of Arizona and The Pennsylvania State University; thus the scope of the measurements was somewhat limited by available equipment and resources. Nevertheless, by combining capabilities, it was possible to obtain a series of both comprehensive and unique lidar and aircraft measurements. The in situ aircraft observations were made as close as possible in time and space to the region(s) probed remotely by the lidar system(s). Following the field experiments in November 1974, each group then independently analyzed its respective measurements. Only after the independent analyses were complete were results intercompared and then attempts made to resolve the few observed differences between the data sets.

2. Sensing techniques and instrumentation

a. Aircraft system

1) Aircraft instrumentation

The Penn State research aircraft, an Aerocommander 680E, is equipped with a variety of meteorological state parameter and winds, turbulence, aerosol and solar infrared radiation sensors. For the Tucson studies (Fig. 1 and Table 1) the following continuous aerosol samplers were used: optical particle counter (OPC), Royco Model 225; integrating nephelometer, MRI
Model 1550; particle mass monitor (PMM), Thermosystems Model 3200A; and a condensation nucleus counter (CNC), Environment One Model Rich 100. Direct aerosol samples were obtained using Unico and Casella cascade impactors and a polystyrene filter sampler. In the Casella impactor electron microscope copper grids coated with carbon film were used as the collecting surface. Both scanning and transmission

TABLE 1. Summary of remote and in situ measurements.

<table>
<thead>
<tr>
<th>University of Arizona multi-wavelength solar radiometer</th>
<th>University of Arizona monostatic lidar</th>
<th>University of Arizona bistatic lidar</th>
<th>Pennsylvania State instrumented aircraft</th>
</tr>
</thead>
<tbody>
<tr>
<td>Optical depth</td>
<td>Backscatter coefficient ($\beta$)</td>
<td>Scattering matrix elements</td>
<td>State parameters</td>
</tr>
<tr>
<td>(at 8 wavelengths)</td>
<td>Extinction coefficient ($\alpha$)</td>
<td></td>
<td>Wind</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>White light scatter coefficient</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Size distribution</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Condensation nuclei</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Mass concentration</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Particle shape</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Particle chemistry</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Radiation fluxes*</td>
</tr>
</tbody>
</table>

B. Derived particulate parameters

<table>
<thead>
<tr>
<th>Columnar</th>
<th>Vertical profile</th>
<th>Selected height</th>
<th>Columnar, selected height and vertical profiles</th>
</tr>
</thead>
<tbody>
<tr>
<td>Size distribution</td>
<td>Extinction coefficient</td>
<td>Size distribution</td>
<td>Extinction coefficient</td>
</tr>
<tr>
<td>Mass loading</td>
<td>Extinction coefficient to backscatter coefficient ratio $S$</td>
<td>Complex particle refractive index</td>
<td>Size distribution</td>
</tr>
<tr>
<td></td>
<td>Mass loading</td>
<td>Number density</td>
<td>Mass loading</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mass loading</td>
<td>Size dependent shape and chemistry</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Number density of some particulate</td>
</tr>
</tbody>
</table>

* Infrared and three visible cutoff filters.
electron microscopes were then used for size distribution, shape and chemical-size distribution analyses.

The concentration and size (in four ranges) of particles ranging from 0.5 to $\sim 8 \mu m$ diameter were obtained using the Royco OPC (Williams and Hedley, 1972). Four different sizes (0.6, 1.0, 2.0 and 5.7 $\mu m$ diameter) of monodisperse polystyrene latex spheres were used for laboratory calibration. Measurements of the atmospheric scattering coefficient $B_{\text{scat}}$ were made with the integrating nephelometer (Charlson et al., 1967; Rabinoff and Herman, 1973). It was calibrated using both pure air and Freon-12 Rayleigh scatters. In the particle mass monitor particles are charged and collected on a quartz crystal sensor. Particles $>0.01 \mu m$ are collected with high efficiency according to Olin et al. (1971). From the frequency difference as a function of time between it and a similar reference crystal, the particle mass may be estimated. The mass monitor was also calibrated using known sizes and concentrations of monodisperse latex spheres. In the condensation nuclei counter the particles are exposed to supersaturations such that particles of diameter $\geq 0.0025 \mu m$ are activated and can be counted.

To eliminate airborne sampling errors all the aerosol used for sizing analyses was sampled through the iso-kinetic decelerator intake (Pena et al., 1977). Situated above the aircraft well beyond the fuselage boundary layer, the probe decelerates the air for sampling from the aircraft speed ($\sim 70 \text{ m s}^{-1}$) to 4 m s$^{-1}$. To minimize line losses, tubes from the intake to each respective sensor were kept as short as possible—all lengths were probably shorter than those found in most surface laboratory installations. In order to eliminate humidity effects the tubes to the OPC and PMM were heated (variations in excess of 85% are not uncommon in a vertical profile).

On-board meteorological observations included pressure, temperature and dew point. Vector winds were obtained from true air speed, heading and Doppler radar ground speed and drift angle. Aircraft position was available either from integrated Doppler radar or recorded navigational DME and VORTAC data.

All flight data were recorded on a 40-channel digital data logging system at 0.5 s intervals. Thus, for those sensors with sufficiently short time constants, approximately a 35 m horizontal or 2.5 m vertical ($\sim 1000 \text{ ft min}^{-1}$ climb or descent) spatial resolution was achieved. This is more than adequate for “urban scale” aerosol studies (Schere and Thomson, 1975; Mamane and Pena, 1975).

To facilitate probing aerosol layers of interest and to insure coordination between the aircraft maneuvers and lidar firing, continuous air-ground radio communication was maintained.
2) Flight patterns

Flight patterns were designed to maximize information about both the vertical and horizontal distribution of particulates. Each flight included a close spiral vertical profile pattern northwest, normally upwind, of Tucson. At \( \sim 152 \text{ m} \) or \( \sim 305 \text{ m} \) intervals, the spiral was interrupted for 30 s to 2 min to enable the CNC radiometer to stabilize. Vertical profiles normally extended from 0.5 to 3.5 km AGL (above ground level). Horizontal sampling within and in the near vicinity of the lidar common volume consisted of rectangular patterns with legs of 30 to 100 km length.

During the week of 18 November 1974, eight flights of \( 2-3 \text{ h} \) duration were completed. The morning and afternoon flights were coordinated with monostatic lidar probing, evening flights with combined monostatic and bistatic lidar sounding.

As the best coordinated set of lidar and flight data was obtained on 20 November, results from flight 5 made on that day, shown schematically in Fig. 2, are the most thoroughly discussed in this paper. Results obtained for this flight are representative of what was observed for most of the other flights during the experiment.

b. Remote sensing systems

The University of Arizona's remote sensing instrumentation employed in the joint experiment included a combination monostatic/bistatic lidar and a multiwavelength solar radiometer. Each of these systems has been described elsewhere in the literature (Reagan and Herman, 1970, 1972; Shaw et al., 1973). The particulate properties which can be obtained directly or inferred from measurements made with each of these systems are also summarized in Table 1.

1) Solar radiometer

The multi-wavelength solar radiometer measures the directly transmitted solar radiation received at the earth's surface as a function of solar zenith angle at eight quasi-monochromatic wavelengths. Radiant energy within certain selected wavelength intervals is isolated by transmission through relatively narrow-band interference filters (bandpass typically \( \sim 10 \text{ nm} \)) arranged in a rotating filter wheel. Readings are made at a rate of about one filter per minute, thereby requiring 8 min to repeat a measurement at a given wavelength. Solar tracking is accomplished with a passive tracking equatorial mount. Output and coding signals are sampled and digitized by a digital voltmeter, and the digital data are recorded on punched paper tape. The entire system is automated to the extent that the radiometer may be left unattended throughout most of the day.

As described by Shaw et al. (1973), the total atmospheric optical depth \( \tau_\lambda(\lambda) \) at each radiometer filter wavelength \( \lambda \) may be obtained from Langley plots of the radiometer measurements for each filter. The total optical depths may also be determined by an intercept method if the radiometer zero airmass intercepts are accurately predetermined for each filter. Both methods were used to analyze radiometer data collected during the joint experiment.

The total optical depth \( \tau_\lambda(\lambda) \) includes components due to molecular (Rayleigh) scattering \( \tau_R(\lambda) \), molecular absorption (due essentially to ozone for the radiometer wavelengths) \( \tau_\sigma(\lambda) \), and particulates \( \tau_p(\lambda) \), the latter being the component of interest. The molecular optical depth components may be determined fairly accurately from theoretical scattering calculations and published norms for columnar ozone amounts, thereby permitting \( \tau_p(\lambda) \) to be obtained by subtracting the computed value of \( \tau_R(\lambda) \) and \( \tau_\sigma(\lambda) \) from \( \tau_\lambda(\lambda) \). The particulate optical depth values are in turn used to infer the particulate columnar size distribution and columnar mass loading. In particular, an effective Junge size distribution fit may be determined by a least-squares analysis of the \( \log \tau_p(\lambda) \) vs \( \log \lambda \) curve (Shaw et al., 1973). Alternatively, the particulate size distribution may be extracted by mathematically inverting the \( \tau_p(\lambda) \) values by techniques similar to those employed by Yamamoto and Tanaka (1969). Once the size distribution is inferred by either of these methods, the columnar particulate mass loading may be readily determined with only the additional specification of the density of the particulates.

2) Monostatic lidar

As described in numerous review articles (e.g., Collis, 1970; Hall, 1974), monostatic lidar measures the fraction of transmitted light backscattered by the atmosphere as a function of range. The University of Arizona monostatic lidar utilizes a 1 J Q-switched ruby laser (wavelength 694.3 nm) for the transmitter, and the receiver consists of an 8-inch telescope-pinhole assembly mounted alongside the transmitter. The photomultiplier output is recorded using a digital data acquisition system consisting of a custom gain-switching amplifier (Spinhirne and Reagan, 1976), Biomation 610B transient recorder, and a paper tape punch. An energy monitor is included that provides an accurate reference of the laser output energy (Reagan et al., 1976), and the lidar is calibrated via the standard target method described by Hall and Ageno (1970).

The monostatic lidar observations made during the joint experiment were composed of sets of slant path measurements. The lidar was fired along a fixed azimuth at several slant angles ranging from vertical decreasing to about 15° above the horizon. A complete data run consisted of measurements at 10–15° angles and required about 30–40 min to complete. Multiple laser shots
were made at each angle to average out the shot noise present on individual returns.

By employing the slant-path data reduction techniques described by Fernald et al. (1972) and Spinhirne et al. (1975), considerable information about atmospheric particulates may be extracted from single-wavelength, slant-path lidar measurements. In particular, slant-path measurements made with a calibrated lidar system can be used to determine absolute values of the particulate unit volume backscattering and extinction coefficients as a function of height. These quantities may be combined with the solar radiometer and bistatic lidar data to aid in the determination of the particulate size distribution, particle refractive index and mass loading.

3) BISTATIC LIDAR

Bistatic lidar operation parallels that of monostatic lidar, except that the receiver and transmitter are spatially separated to permit scattering measurements to be made at various scattering angles. The University of Arizona bistatic lidar system utilizes the same transmitter as the monostatic system. An 8-inch telescope-photonmultiplier assembly spaced 9.52 km from the transmitter serves as the bistatic receiver. The transmitter and bistatic receiver are each mounted on accurate altitude-azimuth positioning mounts to permit rapid and accurate selection of different scattering angles. Bistatic data runs conducted for the joint experiment consisted of horizontal scans covering about 10 scattering angles between 120° and 160°. The time required to complete a scan typically ranged from 60 to 90 min. Simultaneous monostatic lidar measurements were collected during the bistatic scans, and these measurements were used to scale out horizontal particulate inhomogeneities.

The basic parameters which are sought from the bistatic measurements are what are typically referred to as the unit volume particulate scattering matrix, or phase matrix, elements. They characterize the polarization and amplitude scattering properties of particulates in the bistatic common volume for scattering through a given scattering angle. The angular scattering patterns of these parameters are used to infer the particulate size distribution and refractive index by model fitting and inversion procedures similar to those applied to the solar radiometer optical depth versus wavelength data. The reduction procedure for extracting the particulate scattering matrix element values from the bistatic measurements is rather lengthy and requires supporting data from the monostatic lidar and solar radiometer measurements. Details concerning the bistatic data reduction steps will not be given here as they are described in other papers (Reagan and Herman, 1970; Reagan et al., 1977).
3. Results

a. General conditions

On the evening of 20 November, the Tucson area was under the influence of a high-pressure system. Light westerly winds were observed aloft and light easterly winds at the surface. High and middle clouds covering the sky early in the day cleared during the course of the afternoon.

Two temperature inversions (Fig. 3) at the top of (\(\sim 2.4 \text{ km AGL} \)) and slightly above (\(\sim 2.6 \text{ km AGL} \)) the mixing layer were observed on the descending aircraft vertical profile between 1815 and 1855 MST. Within the mixing layer below the inversion, the temperature lapse rate was highly uniform and nearly adiabatic. The relative humidity within the mixing layer increased nearly monotonically from a minimum of \(\sim 30\% \) to a maximum immediately below the inversion of \(\sim 70\% \). As is often observed, the relative humidity dropped sharply, to less than 10\%, above the inversion.

Within the mixing layer the particulate concentration was nearly height independent to 2.4 km as shown by the OPC profile measurements given in Fig. 4. Had it been possible to extend the aircraft profile to the surface, higher concentrations would almost certainly have been found in its immediate vicinity. A decrease in concentration at the top of the mixing layer of approximately one to two orders of magnitude is an often observed feature (Schere and Thompson, 1975; Mamane and Pena, 1975).

Results of OPC size distribution measurements at 10 different heights are given in Fig. 5. The size distributions are similar in shape but differ significantly in number concentrations below and above the mixing layer in agreement with the particle concentration profile given earlier (Fig. 4). Because of the very low concentration of the largest particles, sampling error may have contributed to the variability in size range V. Fewer particles are present in the smallest size ranges than would have been predicted on the basis of a simple "Junge" model fit.

Horizontal flights made at four different heights within the mixing layer indicated fairly good horizontal homogeneity in the particulate properties. Pertinent results are summarized in Table 2 and Figs. 6 and 7. Averages obtained at each level were in general agreement with the earlier vertical profiles. At a given height, the standard deviation of a measured parameter such as particle concentration was typically 10–15\% of the mean, but standard deviations as large as 25\% were observed. On the whole, the assumption of horizontal homogeneity for analysis of the lidar data appeared to be fairly well justified for these particular observations. However, ample evidence exists from both indirect sounding (Russell et al., 1974) and airborne measurements (Godowitch, 1976) that such an assumption is by no means always justifiable and may lead to fallacious results.
Table 2. Summary of the horizontal sections of Flight No. 5 on 20 November 1974.

<table>
<thead>
<tr>
<th>Height</th>
<th>0.53 km</th>
<th>0.88 km</th>
<th>1.03 km</th>
<th>1.18 km</th>
</tr>
</thead>
<tbody>
<tr>
<td>Path</td>
<td>rectangle</td>
<td>square</td>
<td>square</td>
<td>square</td>
</tr>
<tr>
<td>RH (%)</td>
<td>29.9 ±0.8</td>
<td>35.4 ±0.7</td>
<td>38.2 ±0.4</td>
<td>41.0 ±0.06</td>
</tr>
<tr>
<td>$B_{\text{est}}$ (km$^{-1}$)</td>
<td>0.034±0.003</td>
<td>0.032±0.003</td>
<td>0.031±0.004</td>
<td>0.014±0.008</td>
</tr>
<tr>
<td>LPC (cm$^3$)</td>
<td>1.17 ±0.12</td>
<td>1.24 ±0.15</td>
<td>0.96 ±0.18</td>
<td>0.84 ±0.22</td>
</tr>
<tr>
<td>Mass concentration (µg m$^{-3}$)</td>
<td>12.5 ±1.9</td>
<td>9.0 ±1.4</td>
<td>not available</td>
<td>not available</td>
</tr>
</tbody>
</table>

LPC = Large Particle Concentration; $B_{\text{est}}$ = scattering coefficient.

b. Columnar measurements

Due to cloud cover during most of the day, radiometer measurements were not made on 20 November. However, observations were made on both 19 and 21 November. In order to obtain the best possible estimates for the 20th, the intercept method was used to calculate optical depths for the afternoon of 19 November and the morning of 21 November. The particulate optical depths $\tau_p(\lambda)$ determined for these periods are listed in Table 3. Errors included in the table are estimates of the composite errors due to uncertainty in the radiometer zero-air mass calibration constant, atmospheric fluctuations, and uncertainty in the ozone absorption (which is particularly large for the 521.7 and 612 nm wavelengths). The values for each day are highly similar and, we believe, provide a reasonable estimate for the intervening day, 20 November.

The particulate optical depths in Table 3 were used to infer the columnar particulate size distribution and mass loading. A Junge size distribution fit to the data was obtained by performing an error-weighted, least-squares straight-line fit to the log $\tau_p(\lambda)$ vs log $\lambda$ curve. This is based on the assumption that

$$\tau_p(\lambda) = K \lambda^{-\nu + 2},$$

where $\nu$ is the Junge power law parameter and $K$ a wavelength-independent term which varies with particle

![TUCSON, FLIGHT #5
20 NOV. 1974.
1855–1930, MST
HORIZONTAL RUN](image)

Fig. 7. Variations in total particle concentration observed during an extended horizontal flight leg.
Table 3. Particulate optical depths extracted from solar radiometer data collected in Tucson.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>440.0</td>
<td>0.104±0.003</td>
<td>0.100±0.005</td>
</tr>
<tr>
<td>321.7</td>
<td>0.044±0.008</td>
<td>0.044±0.008</td>
</tr>
<tr>
<td>612.0</td>
<td>0.031±0.018</td>
<td>0.035±0.017</td>
</tr>
<tr>
<td>712.0</td>
<td>0.045±0.004</td>
<td>0.040±0.003</td>
</tr>
<tr>
<td>797.7</td>
<td>0.034±0.003</td>
<td>0.029±0.002</td>
</tr>
<tr>
<td>871.7</td>
<td>0.030±0.002</td>
<td>0.037±0.003</td>
</tr>
<tr>
<td>1030.3</td>
<td>0.031±0.003</td>
<td>0.037±0.003</td>
</tr>
</tbody>
</table>

Refractive index. Junge size distributions of spherical particles are reasonably well-approximated by this relation as long as the particle radius range extends from a few hundredths of a micron to several microns (Shaw et al., 1973). After obtaining an estimate of \( \nu \), the particulate column mass loading ML was inferred by the relation (Shaw et al., 1973)

\[
ML = \frac{\Gamma_0 \tau_p(\lambda_0)}{\sigma_p(\lambda_0, \nu)},
\]

where \( \tau_p(\lambda_0) \) is one of the radiometer-derived particulate optical depths for filter wavelength \( \lambda_0 \), \( \sigma_p(\lambda_0, \nu) \) is a theoretically computed extinction coefficient\(^3\) for \( \tau_0 \) and \( \nu \) (and for an assumed refractive index), and \( \Gamma_0 \) is the specific mass per unit volume for which \( \sigma_p(\lambda_0, \nu) \) was computed. The \( \nu \) and ML values obtained by these procedures for the data in Table 3 are given in Table 4. The errors in \( \nu \) and ML are rms estimates due to both errors in \( \tau_p(\lambda) \) and the least-squares straight-line fit used to determine \( \nu \). Mass loading determinations are given for two sets of particle refractive index \( n \) and material density \( \rho \) assumptions, viz., \( n = 1.54–0.0005i \) and \( \rho = 2 \) g cm\(^{-3} \) for “dry particles” (like silicates) and \( n = 1.4–0.0005i \) and \( \rho = 1.4 \) g cm\(^{-3} \) for “wet particles” (such as H\(_2\)SO\(_4\)–H\(_2\)O-type particles with about 50% H\(_2\)O). By a fortuitous cancellation, both sets of particle properties yield about the same columnar mass loading values for the specified \( \nu \) values. The dry particle properties are representative of what is frequently assumed or inferred for naturally occurring atmospheric particulates in the southwestern United States (e.g., see Grams et al., 1974). In contrast, selection of the wet particle properties was stimulated by 1) the abundance of liquid or liquid coated particles collected on the microscope grids in the aircraft, and 2) the apparent best fit of the bistatic lidar data by a refractive index that is characteristic of particles with these properties. Measured particle properties are discussed further in later sections of this paper.

c. Profile measurements

An extended series of particulate backscatter and extinction profiles were measured using the monostatic lidar during the week of the joint experiment. Depending on the prevailing meteorological conditions, the profiles varied significantly from day to day and occasionally even hour to hour. As observed in other lidar/meteorological experiments (Reagan, 1968a, b; Thomson, 1968; Fernald et al., 1972; Utte and Russell, 1974), pronounced scattering inhomogeneities in the lidar profiles were highly correlated with features in the vertical temperature and humidity profiles. For example, lidar backscatter profiles collected coincidently with our aircraft temperature and humidity soundings in Fig. 3 revealed a fairly well-mixed particulate extending up to the temperature inversions at about 2.5 km.

The particulate extinction profiles measured by the airborne integrating nephelometer and the monostatic lidar for the evening of 20 November are shown in Fig. 8. Fixed-height extinction values obtained from horizontal aircraft flights (nephelometer) and bistatic lidar measurements are also included for comparison. About 40 min were required for the descending aircraft profile. The large apparent noise in the nephelometer output above the mixing layer (\( z > 2.5 \) km) and probable significant absolute error are the result of it operating near threshold detection levels (i.e., <5% of full scale and especially at noise level for \( z > 2.5 \) km). The lidar profile was determined from a 90 shot, 5 All of the particulate optical scattering and extinction computations used in analyzing the radiometer and lidar measurements are determined from Mie theory which only applies to spherical particles. It is known that both naturally occurring and man-made particles are not perfectly spherical in shape, but the Mie theory offers the only tractable approach to computing the scattering properties of randomly oriented arrays of randomly shaped particles. Experimental work by Holland and Draper (1967) and Holland and Gagne (1970) indicates that there can be rather large departures of the measured particle scattering parameters from those computed for spherical particles. On the other hand, Eiden (1966), Bullrich et al. (1969) and others have used the spherical assumption and obtained good agreement between theory and observation in studies of the scattering by atmospheric particulates.

\(^3\) The \( \sigma_p(\lambda, \nu) \) values were computed by numerically integrating the Mie single particle extinction cross sections over an assumed particle radius range extending from 0.02 to 10.0 \( \mu \)m, and the integration radius increment \( \Delta r \) was chosen so that the size parameter increment, \( \Delta \Lambda = 2 \Delta r / \lambda \), was less than 0.2. These integration requirements were also applied to the computation of the unit volume backscattering and angular scattering coefficients used in analyzing the lidar data.

Table 4. Size distribution and mass loading estimates inferred from particulate optical depth data.

<table>
<thead>
<tr>
<th>Date</th>
<th>Time (MST)</th>
<th>Junde</th>
<th>( n = 1.54 )</th>
<th>( \rho = 2 ) g cm(^{-3} )</th>
<th>Columnar mass loading (ML) (g m(^{-2} ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>19 November 1974</td>
<td>1601–1644</td>
<td>3.73±0.11</td>
<td>0.058±0.015</td>
<td>0.057±0.015</td>
<td>0.008±0.003</td>
</tr>
<tr>
<td>21 November 1974</td>
<td>0804–0901</td>
<td>3.43±0.05</td>
<td>0.049±0.008</td>
<td>0.044±0.008</td>
<td>0.008±0.008</td>
</tr>
</tbody>
</table>
10 angle slant-path run made over approximately a 35 min interval. Sufficient shots were averaged at each slant-path angle to make the shot noise contribution comparatively unimportant. Error in the lidar extinction coefficient determination (~18% rms error within mixing layer) is therefore attributed primarily to horizontal inhomogeneity and uncertainty in the lidar system's calibration coefficient.

It is evident that the nephelometer and lidar particulate extinction profiles are in close agreement on at least a relative basis (linear correlation coefficient \( r = 0.88 \)). Absolute values cannot be intercompared because the lidar determination is monochromatic (at 694.3 nm), whereas the nephelometer determination is broadband (~100 nm spread) and centered about a significantly different wavelength (500 nm). As an approximation, one can treat the nephelometer reading as approximately monochromatic and scale the lidar extinction coefficient to the center wavelength of the nephelometer. Assuming a Junge-type size distribution which extends over a broad size range, the particulate extinction coefficient \( \sigma_p(\lambda) \) will have approximately a \( \lambda^{-6.6} \) wavelength-dependence [like the optical depth wavelength dependence given earlier in Eq. (1)]. Using the radiometer-derived value of \( \nu = 3.43 \) in Table 4 from the closest in time available radiometer measurements, and the average value of the lidar \( \sigma_p \) between about 0.6 and 2.3 km [\( \sigma_{p,\text{avg}}(\lambda_{\text{lidar}}) = 0.019 \text{ km}^{-1} \)], the scaled value of \( \sigma_p \) at \( \lambda = 500 \text{ nm} \) assuming a \( \lambda^{-6.6} \) dependence is \( \sigma_{p,\text{scaled}} (500 \text{ nm}) = 0.030 \text{ km}^{-1} \). This value comes remarkably close, only about 15% below, the nephelometer value of 0.035 \text{ km}^{-1} averaged over the same height range.

It should also be noted that the lidar extinction profile in Fig. 8 is extremely well correlated with the large particle concentration profile given earlier as verified by a linear correlation coefficient of \( r = 0.92 \). The high correlation indicates very little change in the shape of the particulate size distribution and particle refractive index with height through the mixing layer. This, in turn, means that the S ratio of particulate extinction divided by particulate backscatter, \( S = \sigma_p / \beta_p \), should indeed be nearly constant with height, which is a basic constraint assumed in the lidar slant-path analysis.

The particulate mass concentration profiles measured directly by the aircraft and inferred from the monostatic lidar measurements (with the support of the radiometer data) are given in Fig. 9. The fixed-height mass concentration derived from the bistatic lidar size distribution...
determination is also included for comparison. Determination of the monostatic lidar mass profile is based on the same relation used for obtaining radiometer columnar mass loading [Eq. (2)] except that the lidar particulate extinction coefficient $\sigma_p(\lambda)$ at a given height $z$ is substituted for particulate optical depth $\tau_p$ to give a mass per unit volume at $z$ rather than a columnar value. The lidar profile was computed for $\nu=3.43$ (the value inferred from the closest radiometer measurements) and for wet particle characteristics. The dry particle case is not included because, like the solar radiometer results in Table 4, the values for the two sets of particle characteristics are almost the same. The rms error included in the lidar mass profile is due only to the error in $\sigma_p$. Additional uncertainty in the lidar mass determination obviously arises if the particle size distribution is non-Junge, if the assumed particle density is incorrect, or if the assumed particle size range is incorrect. It is difficult to assess the magnitude of these effects, but they could easily modify the mass by a factor of 2. The fact that the monostatic lidar profile agrees very well with the bistatic lidar fixed-height determination suggests that the size distribution and size range assumptions made for the monostatic analysis were acceptable for at least this occasion.

The aircraft measurements fall below the lidar mass profile with the difference being greatest for the mixing layer where the aircraft (average) value is $12.0\pm1.8$ $\mu g$ m$^{-3}$ and the monostatic lidar height-averaged value is $20.0\pm3.6$ $\mu g$ m$^{-3}$. One very plausible reason for the aircraft value to fall below the lidar value is that wet particles collected by the aircraft were dried in the sampling process and thereby reduced in mass. For example, the average aircraft value for the mixing layer of $12.0\pm1.8$ $\mu g$ m$^{-3}$ does scale to an effective in situ value of $14.3\pm2.2$ $\mu g$ m$^{-3}$, which is closer to the lidar value, if one assumes that 1) 30% of particulates are initially $H_2SO_4\cdot\delta H_2O$ droplets at the in situ humidity, and 2) sufficient water was removed from the droplets to increase the concentration to that required for equilibrium vapor pressure at the humidity in the mass monitor ($\sim10$% RH). Assumption 1 was made because the electron microscope analysis (discussed in the following section) revealed at least 30% of the sampled particles were sulfuric acid droplets. That the effective in situ value is still about 28% below the lidar value (although the rms error limits of the two quantities do overlap) could be due to 1) error in the particle density assumed for the lidar determination, 2) somewhat more than 30% of the particles actually being sulfuric acid droplets, and 3) particle collection losses in the aircraft sampling system. In any case, it is clear that correcting for the drying effect in the aircraft sampling process yields a scaled value in favorable agreement with the monostatic lidar determination.

As a final profile comparison, the aircraft mass measurements were height-integrated and interpolated to surface based high-volume sampler measurements in order to estimate a columnar value which could be contrasted with the radiometer columnar mass loading determination. The estimate made the following assumptions: A $0.7$ $\mu g$ m$^{-3}$ mass concentration was assumed from 2.4 to 12.4 km, the tropopause height, and the stratospheric contribution was assumed to be negligible. The mixing-layer value of $12.0$ $\mu g$ m$^{-3}$ given earlier was applied over the height range from 0.4 to 2.4 km. Finally, the mass concentration was assumed to decrease exponentially from 1 to 400 m, starting with a 1 m value of $91.8$ $\mu g$ m$^{-3}$ [based on high-volume sampler measurements at Tucson by Moyers, (private communication)] and falling to the $12.0$ $\mu g$ m$^{-3}$ at 400 m. The resulting columnar mass was found to be 0.044 g m$^{-2}$. If the effective in situ value of $14.3$ $\mu g$ m$^{-3}$ determined earlier is assumed to apply from 0.4 to 2.4 km, the resulting columnar mass is 0.049 g m$^{-2}$. Either of these values agree favorably with the radiometer values given earlier in Table 4.

d. Electron microscope analysis

Carbon-coated copper screen grids (200 and 400 mesh) were mounted in a Casella cascade impactor

\footnote{Although the number of particles >0.5 $\mu m$ is approximately constant throughout the mixing layer, we have often observed (using the aircraft) that the surface concentration is 5-10 times that of the mixing layer. The characteristics of the surface layer concentration gradient have not been adequately documented due to the difficulty of airborne or instrumented tower sampling near the ground under varied meteorological conditions and surface aerosol concentrations.}
for sampling particles which could then be subjected to electron microscope (EM) analysis. Impactor sampling runs consisted of horizontal (normally "box" pattern flights) lasting 30–45 min (sampling volume, $5.3 \times 10^6$ cm$^3$). Using the transmission EM, micrographs were prepared for particle size distribution and shape analysis, and the scanning EM was used for limited quantitative analysis.

Very few large particles ($>3–5 \mu m$) were deposited on the first two impactor stages. The discrimination between size groups on the various impactor stages is not absolute because the impactor depends upon inertial separation. Thus, larger particles of lesser density may appear on higher order stages, while some smaller high-density particles are deposited on the first or intermediate stages. During the Tucson experiments...
most particulates were deposited on the third and fourth stages.

Figs. 10 and 11 are samples of particles collected on the fourth stage during flight 5 at 530 m AGL. The particle counts follow a log-normal distribution with a mean geometric diameter \( d_g \) of 0.34 \( \mu \)m and a geometric standard deviation \( \sigma_g \) of 2.06. This gives a mean mass diameter of 1.6 \( \mu \)m in good agreement with the value 1.3 given by Renoux and Madelanie (1974). The corresponding mean diameter is 0.44 \( \mu \)m. Fig. 12 shows the distribution of particle types among four different classes (clearly, any number of application-oriented classification schemes are possible). About 30\% of the particles have the characteristic morphology associated with sulfuric acid droplets (Cadle, 1975). Fig. 13 is an example of an apparently extremely large concentration of sulfuric acid type droplets obtained near a smelter in the Tucson area on flight 2. Airborne visual observations suggested that such sources may have significantly contributed to sulfuric acid particulate concentrations observed on this occasion. Non-
``sulfuric acid'' particulates (Fig. 12) included (i) crystal types, \( \sim 5\% \), (ii) porous types (unstable under the EM beam) \( \sim 15\% \), and (iii) others (mostly spheres and stable under EM beam), \( \sim 50\% \).

Particles collected on both the EM grids and the Unico impactor gelatin coated slides were sized and counted both to crosscheck the OPC results and to provide particle sizing information at diameters <0.5 \( \mu \)m. The counts obtained from the EM photographs were corrected for the efficiency of collection of the impactor according to Stern et al. (1962). The composite impactor-OPC size distribution is shown in Fig. 14. Recognizing the problems of droplet spread, particle evaporation on the EM grid and the ambiguity of the OPC in the smaller size ranges (Cooke and Kerker, 1975; van Buijtenen and Oererb, 1974), the agreement between the OPC and impactor results is excellent.

The EM grids were also analyzed with a scanning electron microscope (SEM) using new quantitative methods for the characterization of small particles (0.1–100 \( \mu \)m) (Lebiedzik et al., 1973). In the analysis conducted to date, we have found that large particles (\( \geq 1 \mu \text{m} \)) contain more silicon than sulfur, while the opposite is true for smaller particles:

\[
\begin{align*}
\text{Large particles (} &> 1 \mu \text{m diameter)} \\
\text{Si} &\quad = 5.3 \\
\text{S} &
\end{align*}
\]

\[
\begin{align*}
\text{Smaller particles} &\quad \frac{\text{Si}}{\text{S}} = 0.4.
\end{align*}
\]

This appears to be in agreement with the sulfuric-acid-like droplets found on stages 3 and 4 of the impactor. The few larger crystals observed (which may be silicates) were found mostly on stages 2 and 3. Cunningham and Johnson (1976) observed a similar distribution in aerosols collected in the Chicago and St. Louis areas.

Particle sphericity has also been examined using the SEM. Analysis to date indicates that 50 out of 450 particles could be described as elliptical, with a ratio of major to minor axis \( \geq 2.0 \). The remaining 400 could be well shape-fitted by a circle of the appropriate radius.

e. Fixed-height lidar measurements

Fixed-height bistatic lidar measurements were made at 0.8, 1.6 and 8.0 km AGL on the evening of 20 November. The primary scan, which included measurements at 11 scattering angles, was made at 0.8 km. These were the observations used for inferring the particulate size distribution and refractive index.

At each scattering angle, measurements were made of the fluxes scattered into the bistatic receiver in the planes parallel and perpendicular to the scattering plane (plane of the transmitter and receiver axes) in order to determine the \( l \) and \( r \) Stokes parameters (Chandrasekhar, 1960; van de Hulst, 1957). These measurements were processed to extract \( F_{ll} \) and \( F_{rr} \), the unit volume particulate scattering matrix elements which characterize the particulate scattering influence on the \( l \) and \( r \) scattered Stokes parameters, respectively [See Reagan and Herman (1970) and Reagan et al., 1977 for additional details on data reduction procedures]. The particulate polarization ratio \( R_{rr} \), defined by \( R_{rr} = F_{rr}/F_{ll} \), was the parameter initially
used in the size distribution determination. A plot of $R_{tp}$ versus scattering angle $\theta$ for the 0.8 km data is given in Fig. 15. Each data point is based on the average of several laser shots (typically $\sim 20$), and the error bars denote the estimated rms error due to calibration, shot noise and temporal fluctuations.

The results in Fig. 15 were first compared with families of theoretically computed $R_{tp}$ curves for Junge size distributions with $\nu$ values ranging from 2 to 4 and various refractive index values (real parts of 1.33, 1.40, 1.45, 1.50 and 1.54; imaginary parts of 0.000, 0.002, 0.005, 0.007, 0.010, 0.020 and 0.050). Of these various comparisons, the curves for a purely real refractive index of $n=1.40$ and $\nu$ between 3.0 and 3.6 (curves are included in Fig. 15) agreed best with the data, but they by no means constitute a really good or final fit. Rather, the comparison results were interpreted as indicating 1) the particle refractive index was reasonably well approximated by $n=1.40-i0.000$, and 2) the particle size distribution did not follow a simple Junge model. The refractive index inference is in almost perfect agreement with the refractive index of H$_2$SO$_4$,$\cdot$H$_2$O droplets at the in situ humidity measured at 0.8 km (for $\sim 35\%$ RH, the acid concentration is 50$\%$ for which $n=1.394-i0.000$ at $\lambda=0.7$ $\mu$m, Palmer and Williams, 1975). As the electron microscope analysis indicated that only about 30$\%$ of the sampled particles were H$_2$SO$_4$,$\cdot$H$_2$O droplets, it would seem that the refractive index of the porous and other spherical-type particles (Fig. 12), which account for 65$\%$ of the sampled particles, would also have to be close to $n=1.40$. The refractive index of the crystal type particles is most likely greater than $n=1.50$ ($n \approx 1.54$ at $\lambda=0.7$ $\mu$m for silicates), but the influence of these particles on the effective refractive index value inferred for all particles is minimal in that they comprise only about 5$\%$ of the total particle mix.

Having obtained an estimate of the effective complex refractive index of the particulates ($n=1.40-i0.000$), a library search procedure was then implemented to better fit the bistatic data with a non-Junge size distribution model. The specific model used in the search was the "two-slope" model (Reagan et al., 1972) given by

$$\frac{dN}{dr} = \frac{\left[1 + \left(\frac{r}{r_B}\right)^{\nu+1}\right]}{\left[1 + \left(\frac{r}{r_A}\right)^{\nu+1}\right]} \times D$$

where $dN$ is the number density of particles with radii between $r$ and $r+dr$, $D$ is a scaling parameter, and $r_A$, $\nu_A$, $r_B$ and $\nu_B$ are shaping parameters. The name "two-slope" derives from the fact that the model can define two power-law or Junge-like regions, each of different slope or effective $\nu$ value. The search procedure sought the model parameters which minimized the weighted squared difference between the computed model and measured values of $R_{tp}$, $F_{tp}$ and $F_{trp}$ with the constraint that the model fit also yield $S$ ratio and particulate extinction coefficient $\sigma_p$ values in close agreement with those determined independently from the monostatic lidar measurements. A limited particle size range inference was also made by testing the model parameters for both 0.02 to 5 $\mu$m and 0.02 to 10 $\mu$m size.
ranges and selecting the range which gave better agreement with the measurements. Table 5 gives the best-fit model parameters determined from the search along with various particulate properties given by this set of parameters. The table includes corresponding results for 1) a Junge best fit to the bistatic lidar data using the solar radiometer derived \( \nu \) of 3.43, 2) two Junge fits to the composite aircraft Rich 100-impactor-OPC measurements, and 3) the monostatic lidar "direct" and "indirect" determinations. Plots of these various size distribution determinations are also given in Fig. 14.

In the case of the bistatic Junge fit, the distribution scaling constant \( C \) was selected to minimize the mean-square difference between measured and theoretically computed values of the \( F_{\mu \nu} \) and \( F_{\tau \tau} \) matrix elements. The aircraft Junge fits define maximum and minimum slopes for a compromise power-law fit to the composite Rich 100-impactor-OPC measurements. The Rich 100 only provides a total particle count, but the count may be interpreted as effectively applying to particles with radii \( \gtrsim 0.005 \mu m \), the lower limit due to coagulation, and \( \lesssim 0.1 \mu m \), the radius where the particle concentration typically starts to decrease sharply with increasing particle size (Junge, 1963). In order to provide some estimate of the size distribution in the small particle range, the Rich 100 count was assumed to be due to particles distributed uniformly in concentration (\( dN/ d \log r = \text{constant} \)) over two size ranges, viz., from 0.005 to 0.02 \( \mu m \) and 0.005 to 0.05 \( \mu m \). The smaller slope aircraft power-law fit (for \( \nu = 2.9 \)) was selected to intersect at 0.02 \( \mu m \) with the concentration inferred for the 0.005 to 0.02 \( \mu m \) assumed size range, while the larger slope aircraft fit (for \( \nu = 3.7 \)) was made to intersect at 0.05 \( \mu m \) with the concentration inferred for the 0.005 to 0.05 \( \mu m \) assumed size range. The larger slope aircraft size distribution fit was also extended with a uniform particle concentration from 0.05 \( \mu m \) down to 0.02 \( \mu m \) to span the complete size range of 0.02 to 10.0 \( \mu m \) assumed for calculating the various particle parameters listed in Table 5. Turning now to the monostatic lidar determinations, it should first be recalled that the particulate extinction coefficient \( \sigma_p \) and \( S \) value were determined directly via the slant path technique. The other parameters listed in Table 5 for the monostatic case were calculated using the radiometer inferred Junge \( \nu \) value (\( \nu = 3.43 \)) as well as the \( \sigma_p \) value obtained from the slant path measurements.

4. Discussion

Determination of the particulate size distribution constitutes the most difficult task undertaken in the joint experiment, and this is where the greatest disagreement in the lidar/aircraft results would most likely occur. A comparison of the curves and data
points in Fig. 14 shows that the lidar- and aircraft-derived size distributions are similar in shape, with the aircraft data points typically falling about a factor of 2 lower in radius (for a given concentration) or about an order of magnitude lower in concentration (for a given radius) than the bistatic two-slope distribution. Correspondingly, the extinction coefficient, mass per unit volume and large particle count values given in Table 5 for the aircraft Junge fits are lower, but come within about a factor of \( \frac{1}{2} \) of the lidar measured and inferred values. The level of agreement attained between the lidar and aircraft size distribution determinations, while not extremely close, is certainly not unfavorable in view of the fact that the distributions span about seven orders of magnitude in particle concentration. Moreover, the fact that the aircraft

### Table 5. Intercomparison of particulate parameters measured directly or derived from size distributions inferred from measurements at 0.8 km AGL, Tucson, 20 November 1974.

<table>
<thead>
<tr>
<th>Particulate parameters</th>
<th>Bistatic lidar two-slope fit</th>
<th>Bistatic lidar Junge fit</th>
<th>Junge fits to aircraft Rich 100-OPC-impactor measurements</th>
<th>Monostatic lidar and solar radiometer measurements</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( r = 0.1 \ \mu m )</td>
<td>( r = 0.5 \ \mu m )</td>
<td>( r = 3.4 ) ( r = 3.3 ) ( r = 3.7 ) ( r = 2.9 ) ( r = 3.4 )</td>
<td></td>
</tr>
<tr>
<td>Assumed or inferred size range</td>
<td>0.02 to 5.0 ( \mu m )</td>
<td>0.02 to 10.0 ( \mu m )</td>
<td>0.02 to 10.0 ( \mu m )</td>
<td>0.02 to 10.0 ( \mu m )</td>
</tr>
<tr>
<td>Extinction coefficient</td>
<td>0.022 ( \pm ) 0.002</td>
<td>0.026 ( \pm ) 0.003</td>
<td>0.0043</td>
<td>0.0025</td>
</tr>
<tr>
<td>S ratio</td>
<td>32.1</td>
<td>39.7</td>
<td>39.5</td>
<td>39.7</td>
</tr>
<tr>
<td>Mass per unit volume (( \mu g \ \text{m}^{-2} ))</td>
<td>19.2 ( \pm ) 2</td>
<td>27.4 ( \pm ) 2.7</td>
<td>4.8</td>
<td>1.9</td>
</tr>
<tr>
<td>Large particle count LPC (particles/cm(^{-3} ))</td>
<td>8.8 ( \pm ) 0.9</td>
<td>13.6 ( \pm ) 1.4</td>
<td>2.3</td>
<td>1.2</td>
</tr>
<tr>
<td>( r &gt; 0.25 \ \mu m )</td>
<td>0.73 ( \pm ) 0.07</td>
<td>1.26 ( \pm ) 0.13</td>
<td>0.18</td>
<td>0.17</td>
</tr>
<tr>
<td>( r &gt; 0.5 \ \mu m )</td>
<td>8.9 ( \pm ) 1.6</td>
<td>8.9 ( \pm ) 1.6</td>
<td>0.82 ( \pm ) 0.14</td>
<td></td>
</tr>
</tbody>
</table>

* Two-slope parameters are defined in Eq. (3), and Junge parameters for \( dN/d \log r = Cr^{-n} \).
† The theoretically computed values of \( \sigma_p \) and \( S \) are for a particle refractive index of 1.40 -- 10.000.
‡ The mass per unit volume calculations are for an assumed particle density of 1.4 g cm\(^{-3} \).
determinations are generally smaller than the lidar inferences can be related, at least in part, to specific causes as discussed below.

First, sampling line losses can reduce the number of particles actually delivered to the various sampling devices, particularly for larger particle sizes. For the collection system used in the aircraft, the line losses start to become important for particles with radii $\geq 2$ $\mu$m. In spite of our precautions, it is possible that as much as 50% of the particles with radii $>5$ $\mu$m could have been lost in the sampling lines. Second, the particles delivered to the OPC were first passed through a heater to “dry” hydrosopic type particles. We believe that the drying effect could have reduced the radius of the sulfuric acid type droplets (Fig. 12) by about 30%. Third, the OPC is calibrated with polystyrene latex spheres which have a visible range refractive index of about 1.59–$\pm$0.000, while it was inferred that the effective particle refractive index was about 1.40–$\pm$0.000. (Even accounting for the drying effect on the sulfuric acid type droplets, which comprised about 30% of the particles, their refractive index would only have increased to about 1.43–$\pm$0.000). The exact effect of calibrating the OPC system with particles of significantly larger real refractive index than those actually sampled is difficult to assess without performing light-scattering computations for the specific geometry of the system in question (Royco 225). However, calculations by Cooke and Kerker (1975) for a somewhat similar system (Royco 245) indicate that a refractive index difference of the type inferred here can result in significant error in the inferred radius (by as much as 50% at certain radii) with underestimates in the inferred radius being possible for radii less than about 0.8 $\mu$m. The samples collected by the Casella impactor were not dried during the collection process as there was no heater in the impactor sampling line (Fig. 1), but drying during the transmission EM analysis could have reduced the size of hygroscopic particles. A more likely source of error in the impactor measurements is that of incomplete particle collection due to effects such as particle bounce-off and reentrainment. These effects can significantly impair impactor performance (Dzubay et al., 1976; Natusch and Wallace, 1976), but it is not known how significant these losses may have been during this experiment. Two final considerations which also suggest that the aircraft size distribution measurements were yielding lower than actual in situ particle concentrations are 1) the particle mass monitor readings which (after correcting for particle drying) gave masses close to the lidar determinations; and 2) the integrating nephelometer measurements (the nephelometer sampling line had no heater and was quite short $\sim$30 cm) which, with wavelength correction, gave particulate extinction values in close agreement with the monostatic lidar determinations.

Turning now to possible sources of errors and uncertainties in the remote sensing determinations, it has already been noted that the lidar and solar radiometer inferences include rms error estimates due to measurement errors and temporal fluctuations. Additional factors which influence the remote sensing determinations, but which are not so easily specified, include spatial inhomogeneities, both vertical and horizontal, in the relative shape of the particulate size distribution and in the particle refractive index, uncertainty in the particle refractive index, and the effect of scattering by nonspherical particles. Of these various factors, only spatial inhomogeneities (in relative size distribution shape and particle refractive index) effect the monostatic lidar determination of the particulate extinction coefficient $\sigma_p$ and $S$ ratio.

The aircraft measurements revealed that these inhomogeneity problems were fairly minimal, and slant-path lidar backscatter measurements made simultaneously with the bistatic lidar measurements also indicated good horizontal homogeneity. Thus, the monostatic lidar determinations of $\sigma_p$ and $S$ should be valid within the error limits given for these quantities (Table 5). Moreover, barring significant effects due to nonspherical particle scattering and error in the inferred particle refractive index, the size distribution inferred from the bistatic measurements should produce $\sigma_p$ and $S$ values in agreement with the monostatic lidar determinations. That such agreement was achieved (Table 5) suggests, although by no means constitutes proof, that nonspherical particle scattering apparently had little effect in this case and that the refractive index fit was fairly close. The EM analysis of particles collected by the aircraft impactor also revealed that most of the sample particles, while not perfect spheres, were not greatly nonspherical. With regard to the particle refractive index, the value inferred from the bistatic lidar measurements ($n=1.40–0.000$) was found to be in excellent agreement with at least one of the particle species identified by the EM analysis of the impactor samples (i.e., the sulfuric acid droplets).

Assuming that all of the aforementioned factors which could influence the remote sensing determinations were either unimportant or were properly accounted for, as would appear to be the situation in this case, we now ask, “how closely can the bistatic two-slope best-fit distribution characterize the actual particulate size distribution?” This is difficult to assess because, as with any inversion-type scheme, the effects of measurement errors and limitations in the assumed model and/or applied constraints are not easily separated or defined. One measure of the uncertainty in the two-slope fit is provided by the “envelope” of size distribution curves defined by the various two-slope fits which came within the limits set by the weighted and normalized errors of the parameters used in the search procedure ($\sigma_p$, $S$, $R_{pp}$, $F_{np}$, and $F_{pp}$). On this basis, it was found that the spread in inferred radius for a given concentration was typically no greater than about $\pm 15\%$. 
One of the most notable features of the bistatic two-slope best fit is that it certainly does not follow a simple power law distribution (nor do the aircraft impactor—OPC measurements which define a shape somewhat similar to the bistatic two-slope best fit). While the bistatic Junge fit for $n = 3.43$ does appear to be a fair compromise to the two-slope best fit (Fig. 14), the $S_n$ and $S$ scattering parameter values for this solution deviate much more from the monostatic measurements than the corresponding values for the two-slope best fit (Table 5). The difference is such that the Junge fit considerably overestimates attenuation and forward scatter as compared to what would be obtained for the two-slope best fit. Such overestimates could erroneously affect radiative transfer calculations to determine the heating/cooling influence of atmospheric particulates, which is one of the obvious reasons why it is necessary to infer the particulate size distribution with as much detail as possible.

Results from the columnar and profile measurements require little additional discussion because the aircraft and lidar-solar radiometer determinations were generally in excellent agreement. As noted earlier, the lidar-derived particulate extinction coefficient profile was very well correlated with both the aircraft nephelometer and OPC particle concentration measurements. The lidar and nephelometer particulate extinction coefficient determinations also agreed very closely on an absolute basis—after scaling to compensate for differences in wavelength. The aircraft mass concentration measurements did fall well below the lidar-derived mass profile, but the difference was significantly reduced after accounting for particle drying by the heater in the mass monitor sampling line. Moreover, the lidar mass concentration inference should logically yield one of the poorer comparisons with the aircraft measurements in that an assumption had to be made about the particle density in order to complete the lidar determination. Finally, the columnar mass loading values inferred from the solar radiometer measurements were in very good agreement with the height-integrated aircraft mass measurements when an interpolation to a ground-based mass measurement was included.

5. Summary

Direct aircraft and indirect lidar-solar radiometer determinations of atmospheric particulate size and height distributions have been intercompared. Vertical profiles of particulate unit volume extinction inferred by these two measurement approaches were found to be in excellent agreement on both a relative and absolute basis. Lidar inferences of the particulate mass concentration agreed favorably with the aircraft mass monitor measurements. The particulate size distribution determinations were similar in shape and agreed in absolute particle concentrations within an order of magnitude. In general, no serious discrepancies or unexplainable differences were found between the direct and indirect determinations.

Through the combined bistatic-monostatic lidar and solar radiometer measurements, it was possible to extract both the particulate size distribution and complex index of refraction. The refractive index determination of $n = 1.40-0.000$ differed from the range of values that have frequently been assumed or inferred for arid region particulates [i.e., real parts from 1.5 to 1.55 and imaginary parts from 0.005 to 0.01 (Grams et al., 1974; Lindberg and Laude, 1974)]. This was due at least in part to the fact that a large fraction of the particulate sample was composed of sulfuric acid droplets as confirmed by the aircraft impactor measurements. The size distribution determined from the bistatic-monostatic lidar measurements deviated from a simple Junge fit both in terms of the shape of the distribution and the scattering parameters produced by the distribution.

In conclusion, the results obtained from this joint experiment demonstrate, at least under certain conditions, that:

1) Direct aircraft and indirect lidar-solar radiometer measurements can provide very similar estimates of the size and height distributions of atmospheric particulates.

2) Lidar-solar radiometer measurements can independently provide detailed inferences of the particulate size distribution and complex index of refraction.

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