

In situ measurement of the aerosol extinction-to-backscatter ratio at a polluted continental site

Theodore L. Anderson

Joint Institute for the Study of the Atmosphere and Oceans, University of Washington, Seattle

Sarah J. Masonis, David S. Covert, and Robert J. Charlson

Department of Atmospheric Sciences, University of Washington, Seattle

Mark J. Rood

Department of Civil and Environmental Engineering, University of Illinois at Urbana-Champaign

Abstract. The extinction-to-backscatter ratio S is a crucial parameter for quantitative interpretation of lidar data, yet empirical knowledge of S for tropospheric aerosols is extremely limited. Here we review that knowledge and extend it using a recently developed in situ technique that employs a 180... backscatter nephelometer. This technique allows robust quantification of measurement uncertainties and permits correlations with other aerosol and meteorological properties to be explored. During 4 weeks of nearly continuous measurements in central Illinois, S was found to vary over a wide range, confirming previous indications that geographical location by itself is not necessarily a good predictor. The data suggest a modest dependence of S on relative humidity, but this explains only a small portion of the variation. Most variation was associated with changes between two dominant air mass types: rapid transport from the northwest and regional stagnation. The latter category displayed much higher aerosol concentrations and a systematically higher and more tightly constrained range of S . Averages and standard deviations were 64 ± 4 sr for the stagnant category and 40 ± 9 sr for the rapid transport category. Considering the 95% confidence precision uncertainty of the measurements, the difference between these averages is at least 13 sr and could be as large as 35 sr. The wavelength dependence of light scattering, as measured by a conventional nephelometer, is shown to have some discriminatory power with respect to S .

1. Introduction

With its demonstrated ability to map aerosol variations throughout the atmospheric column [Winker *et al.*, 1996], laser radar (lidar) has become a central technology in current strategies for tropospheric aerosol research [Seinfeld *et al.*, 1996]. Its use is complicated, however, by the fact that the lidar signal contains a convolution of two basic optical properties of the aerosol particles: the 180° backscatter coefficient β_p ($\text{m}^{-1} \text{sr}^{-1}$) and the extinction coefficient σ_{ep} (m^{-1}). A quantitative retrieval of either property requires knowledge of their relationship along the laser path [Klett, 1981]. Here we review and extend current knowledge of this relationship, which we express in the usual form of an extinction-to-backscatter ratio, $S = \sigma_{ep} / \beta_p$, with units of steradians. (To make this definition unambiguous, the value of S in the Rayleigh limit is $8\pi/3$.)

Like other aerosol optical properties, S can be calculated from Mie theory. Mie calculations are essential for instrument validation experiments (e.g., Figure 1) and provide guidance to field measurements by suggesting the range of values to be expected and what the controlling factors ought to be. A study by Ackermann [1998] illustrates

the latter point. He used assumptions for aerosol size distribution and refractive index to show that S should vary from about 15 to 75 sr for tropospheric aerosols, with lower values associated with weakly absorbing coarse-mode particles (i.e., sea salt and mineral dust) and higher values associated with small and/or highly absorbing accumulation-mode particles. These Mie calculations were combined with models of aerosol hydration to develop functional relationships between S and ambient relative humidity for several aerosol types.

While valuable, such calculations are inadequate in themselves for two reasons. First, Mie calculations are limited to simple, frequently unrealistic particle morphologies (homogeneous spheres, concentric spheres, and a few variants). Second, this theoretical approach to determining S can never be more accurate than the empirical knowledge of particle size and refractive index (the latter from knowledge of particle chemistry) upon which the calculation is based. For these reasons, direct empirical determinations of S are required.

Several methods have been developed to make this measurement; however, as shown in Table 1, they have been deployed to date in only a handful of investigations of tropospheric aerosols. Most regions of Earth and many major aerosol types have no published data at all, and a statistically significant database exists for only one location: Tucson, Arizona.

Despite its inadequacy in terms of sample size, the existing database does permit three useful insights. First, the measured range of S values (8-95 sr) is in rough agreement with the Mie predictions cited above. Second, measurements during high coarse-mode concentrations tend toward the lower end of this range, while the smaller, pollution-derived particles are associated with higher values of S . Finally, the large range of values measured at several sites indicates that geographical location by itself does not necessarily provide a useful constraint on S .

2. Experimental Description

Measurements were made from August 28 to September 27, 1999, at the Bondville aerosol research station (BND) in central Illinois (40.05°N, 88.37°W, 229 m). This station operates year-round as the polluted continental site in a network of midlatitude stations managed by the National Oceanic and Atmospheric Administration Climate Monitoring and Diagnostics Laboratory. The sampling system and instrumentation have been described in previous reports [Koloutsou-Vakakis *et al.*, 1998, 1999]. Key properties measured are the mass and ionic chemistry of particles smaller than 1 μm dry aerodynamic diameter (submicron), aerosol light absorption, and the humidity dependence of submicron and supermicron aerosol light scattering. Quality control procedures included a weekly leak check of the entire plumbing system by installing a particle filter at the station inlet.

During this experiment we deployed supplemental instruments sufficient to independently determine the

ambient value of S at 532 nm on a continuous basis. Particulate light extinction was measured as the sum of scattering σ_{sp} and absorption σ_{ap} . The three parameters needed to determine S (σ_{sp} , σ_{ap} , and β_p) were measured with an integrating nephelometer (model 3563, TSI, Inc., St. Paul, Minnesota), a particle soot absorption photometer (model PSAP, Radiance Research, Seattle, Washington), and a 180° backscatter nephelometer, respectively. Operation and data reduction procedures for the first two instruments followed published protocols [Anderson and Ogren, 1998; Bond *et al.*, 1999; Anderson *et al.*, 1999] with some modifications noted below. Design and operation of the 180° backscatter nephelometer and a modified TSI nephelometer are described in a previous paper [Doherty *et al.*, 1999]. Results of a laboratory closure experiment with latex spheres are shown in Figure 1 and evaluated below.

Our goal was to measure S (and other optical properties) at ambient relative humidity (RH) 10 m above the ground for eventual comparison to collocated remote measurements by lidar (the subject of a future report). Sample air was drawn from the base of the station's normal inlet (a 10-m vertical pipe) and then routed into a shed, adjacent to the main building. This shed was protected from rain and sunlight and was actively ventilated with outdoor air. To help reduce sample heating, the nephelometers were operated without covers, the power to the model 3563 lamp was reduced from 75 to 40 W, and a large flow of air was directed across this lamp. (Note that the 180° nephelometer uses a 10-mW laser, which does not cause sample heating.) At night, when the surface temperature dropped below the temperature at the 10-m height of the station inlet, the shed was heated slightly. Through these means, the nephelometers were maintained near the ambient 10-m temperature so that the scattering measurements would correspond closely to scattering at ambient RH. The absorption measurement, in contrast, was made at low RH, due to heating within the PSAP. Lacking empirical data, the possible humidity dependence of σ_{ap} was neglected. (This neglect could cause an underestimate of extinction and thus an underestimate of S . However, because absorption is always a small fraction of extinction, these effects will be small. Note that whatever effect absorbing components in the hydrated state have on scattering and backscattering will be accurately measured by our technique. Only the effect on absorption is unknown.)

3. Data Reduction and Uncertainty

All aerosol properties are reported at ambient temperature and pressure and at the relative humidity (RH) inside the 180° nephelometer. We adopt the latter convention because (1) RH inside the 180° nephelometer is a reasonably good proxy for ambient RH at the station inlet, tracking it to within -10% for the portion of data used herein, and (2) this minimizes the uncertainty in S , since no RH adjustment to β_p is required. A small RH adjustment was required for the model 3563 scattering measurements (see below). Both the measurement of RH and the RH adjustment become highly

uncertain near saturation: for this reason, we report data only when the RH of both nephelometers was below 90%.

The determination of S and other optical properties involves a sequence of steps beginning with the raw measurements and including instrument calibrations as well as various adjustments and corrections. For each of these steps we have developed an uncertainty estimate that reflects the potential for error at the 95% confidence level. In most cases, these uncertainties have been quantified by empirical methods, as briefly explained in the following paragraphs.

Noise uncertainty for all optical instruments was determined by measuring filtered air for 25 min, three times a day and following the propagation scheme outlined by *Anderson et al.* [1999]. This is discussed further in section 4.4, below. The inherent accuracy of the calibrated measurements is assigned a constant value based on laboratory closure experiments. We use -7% for both nephelometers [*Anderson et al.*, 1996] (see Figure 1 and below for the 180...nephelometer) and -20% for the PSAP [*Bond et al.*, 1999]. Calibration stability for both nephelometers was assessed by measuring the calibration gases (air and CO_2) 12 times during the campaign. From these measurements, average values and the 95% confidence uncertainties of the offset and slope calibration factors were determined [*Anderson and Ogren*, 1998]. At low aerosol concentrations, offset uncertainty in the β_p measurement was large, causing overall uncertainty in S to exceed 100% for about 10% of the 10-min data points. These cases have been eliminated from the data set, which tends to bias our samples somewhat away from the low concentration conditions.

The inherent accuracy of the 180...nephelometer was assessed through laboratory measurements of conservatively scattering latex spheres across a broad range of accumulation mode sizes. As shown in Figure 1, measured and predicted values are within experimental uncertainties when diameter uncertainty is taken to be -2% (horizontal error bars) and scattering uncertainty for both nephelometers is taken to be -7% (resulting in -10% uncertainty in S ; vertical error bars). Note that latex spheres are unlike atmospheric particles in the sense that they are nearly monodispersed. Mie theory predicts large, size-dependent oscillations in β_p for submicron particles (owing to constructive and destructive interference in the scattered waves) that are not present in total scatter. This causes the extreme size sensitivity of S values seen in Figure 1 and the existence of values well over 100 sr. Both of these features are unrealistic for atmospheric particles (although reproducing these features constitutes a fairly rigorous test of our measurement system.) For the PSAP measurement of σ_{ap} , a precision uncertainty of -6% was applied as well as a scattering correction (and uncertainty) equal to $2 - 1\%$ of σ_{sp} [*Bond et al.*, 1999].

Wavelength adjustments were made under the assumption that σ_{sp} and σ_{ap} vary with $\lambda^{-\alpha}$, where λ is wavelength and α is the ngstr m exponent. The 3563 nephelometer measures at three wavelengths: 450, 550, and 700 nm. We adjusted the 550-nm measurement to 532 nm based on (450/550) and assumed an uncertainty equal to -50% of the adjustment. The PSAP is calibrated to measure σ_{ap} at 550 nm. We adjusted this

to 532 nm by assuming $\tau = 1$ for absorption. This is consistent with both theoretical and field studies [e.g., *Foot and Kilsby*, 1989]. However, since we had no direct measure of the wavelength dependence of absorption, we assumed an uncertainty equal to 100% of the adjustment. Because the 180° nephelometer operates at 532 nm, no wavelength correction was required for β_p .

Correction factors (and corresponding uncertainties) that account for angular nonidealities of the model 3563 nephelometer, derived by *Anderson and Ogren* [1998], were adopted. The corrections are a strong function of particle size, which is reflected in the measured values of β_p . For this experiment, supermicron particles were a minor contributor to light scattering, as shown both in the measured values of β_p (Table 2b) and in direct assessments using, alternately, 10- μm and 1- μm impactors upstream of the nephelometers. For scattering dominated by submicron particles, as herein, the correction factors are modest (<10%) and the corresponding uncertainties are small. A similar situation exists for the 180° nephelometer. This device actually senses light scattered from 176° to 178°. Adjustment to 180° backscatter is accomplished via gas calibration, as discussed by *Doherty et al.* [1999]. Consistent with calculations by *Doherty et al.* [1999], we assume -5% uncertainty associated with the angular nonideality of the 180° nephelometer.

RH adjustments to σ_{sp} were required to account for differences between the RH in the 3563 nephelometer and that in the 180° nephelometer. Because these differences were small, we made the simple assumption that σ_{sp} varies with $(1 - \text{RH})^{-\gamma}$, with γ set to 0.3, and calculated uncertainty by varying γ between 0.1 and 0.5 [*Kasten*, 1969]. Because the dependence of σ_{ap} on RH is not known, we have not applied an adjustment or attempted to estimate a corresponding uncertainty.

Assuming that the various sources of uncertainty are independent, we combined them in quadrature. For averaged quantities, noise uncertainty is assumed to diminish with the square root of sample time. In addition, uncertainty associated with calibration stability is reduced for averages that span two or more calibrations. All other sources of uncertainty are considered to be systematic, such that they do not diminish with averaging.

4. Results and Discussion

4.1. Overview

We will present and discuss only the prominent features of the data set (the full data set is available via anonymous ftp to ftp.atmos.washington.edu, subdirectory /cpo/linc/UW). Tables 2a and 2b and Figure 2 summarize our measurements. Meteorological properties for the entire data set and for the two dominant air mass categories (defined below) are presented in Table 2a. Aerosol optical properties, divided in this same way, are presented in Table 2b.

The intensive optical properties discussed herein are the extinction-to-backscatter ratio,

$$S = \frac{\sigma_{sp}(532) + \sigma_{ap}(532)}{\beta_p(532)} \quad (1)$$

the single scattering albedo,

$$\omega = \frac{\sigma_{sp}(532)}{\sigma_{sp}(532) + \sigma_{ap}(532)} \quad (2)$$

and the ngstr m exponent,

$$\hat{a} = -\frac{\log\{\sigma_{sp}(450)/\sigma_{sp}(700)\}}{\log(450/700)}, \quad (3)$$

where wavelengths are indicated parenthetically in each case.

For each optical property, Table 2b provides information on the average, the uncertainty of the average (both precision and total uncertainty), and the variance (the standard deviation). Precision uncertainty (repeatability) is the sum of instrumental noise, calibration drift, and RH-adjustment uncertainty. As pointed out by *Anderson and Ogren* [1998], this quantity allows measurements taken with the same instruments and protocols but at different times or places to be compared. Here it allows us to evaluate whether differences in the averages between air mass categories are robust in terms of instrumental uncertainties. Information on the robustness of the standard deviation in terms of instrumental noise is also presented in Table 2b, as discussed in section 4.4 below.

In Figure 2 we examine correlations of S with various potentially controlling factors. For clarity, the Bondville data on these plots are from the two dominant air mass categories only. In Figures 2b and 2c, we include additional data from a coastal station in the Pacific Northwest (Cheeka Peak, Washington, 48.3...N]24.6...W480 m), taken with the same instrumentation during the spring of 1998 [*Doherty et al.*, 1999].

4.2. Air Mass Categories

Air mass categories were established by dividing the 4-week campaign into 10 meteorologically distinct episodes of 1 to several days duration, separated by transition periods of 1 to several hours. The separation of episodes was based on (1) air parcel height and travel distance over the 2 days prior to sampling, determined from twice-daily isentropic back trajectories, and (2) local dew point temperature. In terms of these properties, seven of the 10 episodes, comprising most of the total sample time, were found to fall into two categories: rapid transport from the northwest (long NW; 49% of the data) and moist, regional stagnation (stagnant; 26% of the data). To ensure regional representativeness, an episode was assigned to one of these categories only if the afternoon temperature soundings (taken at Lincoln, Illinois, 50 km to the west) indicated that the surface layer was coupled to the boundary layer.

4.3. General Features of the Aerosol Over Central Illinois

Most optical properties show robust differences between the two dominant air mass categories. This can be seen in Table 2b in terms of both measurement uncertainty (compare means plus or minus the precision uncertainty) and measured variance (compare means plus or minus SD). Careful examination of these distinctions yields a number of insights into the nature of the late summertime aerosol over central Illinois. On average, the stagnant category, compared with long NW, is associated with far higher levels of σ_{sp} (a factor of 7) but only slightly higher levels of σ_{ap} (a factor of 1.5). This suggests (1) that there are independent sources for scattering and absorbing aerosol mass, with the latter being more uniformly distributed in space and time, and (2) that the high scattering mass is due to secondary aerosol produced under stagnant conditions while the absorbing mass is from primary sources. During stagnant conditions the intensive properties, ω and S , are significantly higher and are also much more tightly constrained than during the cleaner, long NW conditions. This suggests (3) that a relatively consistent aerosol type dominates the high pollution events in central Illinois while a more variable mixture of aerosol types is present during cleaner conditions.

Aerosol chemical analysis supports this overall picture and shows that the secondary aerosol produced under stagnant conditions is largely sulfate. Mass and inorganic ions were measured with 4-hour, submicron filter samples during most of the campaign. Nitrate was found to be an insignificant portion of aerosol mass during the entire experiment. Sulfate, on the other hand, was always significant, although it formed a highly variable portion of total mass. (Chemically unidentified mass at Bondville is most likely carbonaceous [Koloutsou-Vakakis *et al.*, 1999]). Defining sulfate mass as the sum of SO_4^{2-} and NH_4^+ masses and defining total mass as the gravimetric aerosol weight at 33% RH, the sulfate-to-total mass ratio averaged 0.36 for the long NW category and 0.67 for the stagnant category. Many large, coal-burning power plants are located within 1000 km of Bondville, the average, 2-day travel distance during stagnant conditions (Table 2a). These are known to emit large quantities of SO_2 to the atmosphere (most operate without sulfur emissions controls) and are the likely source of sulfate aerosol during high pollution events. (Emissions are monitored by the Environmental Protection Agency and reported at www.epa.gov/acidrain/edata.html.)

4.4. Variability of S and Potential Controlling Factors

Table 2b indicates a significant contrast in S between the long NW category (40.5 sr) and the stagnant category (64.3 sr). In terms of 95% confidence limits (using the precision uncertainties), the difference in S between categories is 24 – 11 sr. Thus, in terms of the average properties of characteristic air mass types, the Bondville data indicate a range of S of at least 13 sr and perhaps as much as 35 sr.

The data also indicate considerable variation within air mass categories, as evident in Figure 2 and in the standard

deviations given in Table 2b. However, some of the measured variation could be due to instrumental noise. On the basis of extensive measurements of filtered air (thrice daily for 25 min), noise levels for each optical instrument are well known and can be propagated to calculate noise uncertainty for all parameters at 10-min time resolution. To calculate noise for this purpose, we use the standard deviation of the filtered air measurements at 10-min time resolution over the entire campaign. Defined in this way, noise accounts for any instrumental drift over the campaign. This basic noise level is then increased by the square root of the signal magnitude to estimate noise for each 10-min sample. Finally, the average value of this 10-min noise (i.e., averaged over each air mass category) is used to estimate the instrumentally induced portion of measured variation in Table 2b.

If both noise and the ambient quantity being measured are normally distributed, then their standard deviations will add in quadrature to yield the measured standard deviation. That is, the relation between the measured standard deviation SD_m and the true standard deviation SD_t (i.e., what would be measured with a perfect, noise-free instrument) will be

$$SD_t = \sqrt{SD_m^2 - N^2}, \quad (4)$$

where N is the average magnitude of the 1-sigma instrumental noise level, as discussed above. Table 2b shows both SD_m and SD_t , calculated from (4). Also shown is the ratio SD_m/N , which can be used to assess the robustness of SD_t . For most parameters this ratio is high (over 4), indicating that noise is small relative to measured variance and has a small effect on the measured standard deviation (below 10%). When this ratio drops below 2, however (i.e., when estimated noise and measured standard deviation have similar magnitudes), the accuracy of true standard deviation, SD_t , calculated from (4) becomes questionable. For those cases, Table 2b shows SD_t in parentheses. This condition applies to S for each air mass category. Thus we cannot ascribe high confidence to the measured variations in S within air mass categories. This situation clearly calls for improved precision in our measurement technique (the main culprit being calibration instability of the 180; nephelometer.) At the same time, this result emphasizes the importance of monitoring calibration stability via frequent filtered air measurements and gas calibrations.

Even though the standard deviation of S within air mass categories cannot be determined with high accuracy, a strong argument can be made that the observed variations are real. First, the standard deviations of each of the extensive properties from which S is derived (i.e., σ_{sp} , σ_{ap} , and β_p) are robust (with the possible exception of β_p for the long NW category.) Indeed, the contrast in SD_m/N between extensive and intensive properties in Table 2b provides direct evidence that the latter are much more nearly conserved over time than the former. (Another way to show this is to examine the correlations between extensive properties.) Thus quantifying the variations in intensive properties requires considerably more instrumental precision than quantifying the variations in extensive properties. Second, the variability in S evident in Figure 2 is not a result of the 10-min time resolution of

the measurements. This can be shown by autocorrelation analysis of the 10-min data. The lagged autocorrelation coefficient r [Box *et al.*, 1978, p. 63], falls off smoothly and slowly with increasing temporal offset, reaching values of 0.93 and 0.86 for time offsets of 1 and 2 hours, respectively. In other words, nearly all of the variation is associated with timescales longer than 2 hours, and the 10-min time resolution is not artificially generating variation within air mass categories. Finally, there are many individual portions of the time-series data that show statistically robust (95% confidence) changes in S within air mass categories. We can conclude from the above that even though the variance of S cannot be quantified with high accuracy (due to instrumental noise), the observed variations within air mass categories are real and can legitimately be examined for correlations with potentially controlling factors.

Figure 2 explores three factors that might explain some of the variation in S . For clarity, only data from the two dominant air mass categories are shown. Figure 2a displays S as a function of relative humidity, RH. The functional relationship between S and RH predicted by Ackermann [1998] for the continental aerosol type is also shown. It is important to point out that the aerosol at Bondville is quite hygroscopic, as revealed by both chemical measurements and humidity-dependent light scattering measurements. This hygroscopicity is apparent in our own data (recall that our nephelometers operated at near-ambient RH, not controlled RH) when σ_{sp} is plotted against RH (not shown). Despite this hygroscopicity and the large range of RH encompassed by our measurements (20-90%), we find evidence of only a modest variation in S as a function of RH and the amount of the variability in the data explained by the RH dependence is small. Indeed, the regression-derived slope of S versus RH for our data is not statistically significant for either air mass category.

Figure 2b shows the relationship between S and extinction, σ_e . Here we see that high pollution events in central Illinois have a much narrower range of S than the overall data set. For instance, for $\sigma_e > 60 \text{ Mm}^{-1}$, the 70% range of S is 58 to 71 sr (identical with the 70% range for the stagnant category). Interestingly, the continental aerosol at Cheeka Peak, Washington, which represents diluted pollution from the Puget Sound urban/industrial region, manifests a similar range of S (70% between 55 and 75 sr), although at much lower concentrations. This comparison provides an important caution. The data from Bondville imply that extinction levels may provide a useful guide to predicting S within a source region, such as central Illinois. However, for pollution that is advected outside of its source region (and thus diluted), the relationship between extinction and S will necessarily break down. Thus no general relationship between S and σ_e is to be expected.

On the other hand, a plausible argument can be made for a general relationship between S and the wavelength dependence of scattering, ω . This is because both quantities are intensive properties (not dependent on concentration) and both are highly sensitive to the partitioning of aerosol mass between coarse and accumulation modes. Figure 2c

examines whether our data support such a relationship. For the Bondville data the range of S is relatively small (70% between 1.5 and 2.3), reflecting the fact that accumulation-mode particles were always dominant. Thus no consistent relationship between S and ω emerges from the Bondville data taken alone. (The intriguing negative trend in S versus ω for the stagnant category reflects the fact that both quantities vary with RH, but in opposite directions.) Adding the Cheeka Peak data, which include cases of coarse-mode dominated sea-salt aerosol, increases the range of S to values below 0. Within this larger frame, we see that S is systematically lower for coarse-mode dominated aerosols and that ω is a useful guide to this distinction. No useful relationship between S and ω was found for our data. The plot of this correlation is not included.

5. Conclusions

Using an in situ technique that features a recently developed 180° backscatter nephelometer, we have measured the extinction-to-backscatter ratio S at a polluted continental site in central Illinois for 1 month during late summer 1999. These measurements are put in the context of a previous theoretical study and several previous empirical studies that used remote techniques. The in situ method allows accurate quantification of uncertainties and allows variations in S to be correlated with other aerosol and meteorological properties. Measurements were made at near-ambient relative humidity, RH, which ranged from 20% to 90% for the portion of data analyzed. All optical properties (except the wavelength dependence of scattering) are reported at 532-nm wavelength. The full data set (measured values and 95% confidence uncertainties) is publicly available by contacting the authors. Principal findings presented in this report are as follows.

1. Meteorological analysis based on local dew point temperature and back trajectories showed that 75% of the sample time fell into one of two distinct air mass categories: rapid transport from the northwest (long NW) and moist, regional stagnation (stagnant). Comparison of optical properties between these air mass categories suggests (i) that there are independent sources for scattering and absorbing aerosol mass, with the latter being more uniformly distributed in space and time, (ii) that the high scattering mass is due to secondary, mostly sulfate aerosol produced under stagnant conditions while the absorbing mass is from primary sources, and (iii) that a relatively consistent aerosol type dominates the high pollution events in central Illinois while a more variable mixture of aerosol types is present during cleaner conditions. In all cases, optical properties were dominated by submicron, accumulation mode particles.

2. Intensive aerosol optical properties (specifically, the wavelength dependence of scattering σ , single scattering albedo ω , and S) were observed to be much more nearly conserved over time than the extensive properties from which they are calculated. This effect was quantified in terms of the ratio of measured standard deviation to instrumental noise, SD_m/N , which can be calculated in an equivalent

fashion for both extensive and intensive properties. This ratio fell below 2 for S , indicating that the measurement technique was unable to accurately determine the standard deviation of S within air mass categories. However, our measurements do bound the standard deviation of S . The measured values represent upper limits. The lower limits are well above zero, since statistically robust changes in S were observed in individual portions of the time series.

3. Most variation in S was associated with changes between the dominant air mass types. The averages and standard deviations of S were 40 ± 9 for the long NW category and 64 ± 4 for the stagnant category. Considering the 95% confidence precision uncertainty of the measurements, the difference between these averages is at least 13 sr and could be as large as 35 sr.

4. The data suggest a modest dependence of S on relative humidity in line with the predictions of Ackermann [1998], but this dependence is not statistically significant and explains only a minor fraction of the variation.

5. When combined with previous in situ measurements on the Washington coast, in which a significant coarse-mode sea-salt aerosol was present, the data suggest that the wavelength dependence of light scattering, as measured by a conventional nephelometer, has some discriminatory power with respect to S . In particular, S values are systematically low (around 20 sr) when the ngstr m exponent is below 0.5.

Acknowledgments. We gratefully acknowledge dedicated technical support by many individuals during the field campaign, especially Karen Winter, Casey Anderson, and Uli Steidl. This research was funded primarily by the National Aeronautics and Space Administration's Global Aerosol Climatology Project. The Bondville Station is supported by National Oceanic and Atmospheric Administration (NOAA), contract COM NA 96-GP0342. Partial support for this publication came from NOAA via the Joint Institute for the Study of the Atmosphere and Oceans, agreement NA37RJ0198, contribution 765.

References

- Ackermann, J., The extinction-to-backscatter ratio of tropospheric aerosol: A numerical study, *J. Atmos. Oceanic Technol.*, 15, 1043-1050, 1998.
- Anderson, T. L., and J. A. Ogren, Determining aerosol radiative properties using the TSI 3563 integrating nephelometer, *Aerosol Sci. Technol.*, 29, 57-69, 1998.
- Anderson, T. L., et al., Performance characteristics of a high-sensitivity, three-wavelength, total scatter/backscatter nephelometer, *J. Atmos. Oceanic Technol.*, 13, 967-986, 1996.
- Anderson, T. L., D. S. Covert, J. D. Wheeler, J. M. Harris, K. D. Perry, B. E. Trost, D. J. Jaffe, and J. A. Ogren, Aerosol backscatter fraction and single scattering albedo: measured values and uncertainties at a coastal station in the Pacific Northwest, *J. Geophys. Res.*, 104, 26,793-26,807, 1999.
- Ansmann, A., M. Riebesell, U. Wandinger, C. Weitkamp, E. Voss, W. Lahmann, and W. Michaelis, Combined Raman elastic-backscatter lidar for vertical profiling of moisture, aerosol extinction, backscatter, and lidar ratio, *Appl. Phys. B*, 55, 18-28, 1992.
- Ansmann, A., D. Althausen, U. Wandinger, K. Franke, D. Müller, F. Wagner, and J. Heintzenberg, Vertical profiling of the Indian aerosol plume with six-wavelength lidar during INDOEX: A first case study, *Geophys. Res. Lett.*, 27, 963-966, 2000.

- Bond, T. C., T. L. Anderson, and D. Campbell, Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols, *Aerosol Sci. Technol.*, *30*, 582-600, 1999.
- Box, G. E. P., W. G. Hunter, and J. S. Hunter, *Statistics for Experimenters*, p. 63, John Wiley, New York, 1978.
- de Leeuw, G., G. J. Kunz, and C. W. Lamberts, Humidity effects on the backscatter/extinction ratio, *Appl. Opt.*, *25*, 3971-3974, 1986.
- Doherty, S., T. L. Anderson, and R. J. Charlson, Measurement of the lidar ratio for atmospheric aerosols using a 180°-backscatter nephelometer, *Appl. Optics*, *38*, 1823-1832, 1999.
- Ferrare, R. A., S. H. Melfi, D. N. Whiteman, K. D. Evans, and R. Leifer, Raman lidar measurements of aerosol extinction and backscattering, 1, Methods and comparison, *J. Geophys. Res.*, *103*, 19,663-19,672, 1998.
- Foot, J. S., and C. G. Kilsby, Absorption of light by aerosol particles: An intercomparison of techniques and spectral observations, *Atmos. Environ.*, *23*, 489-495, 1989.
- Kasten, F., Visibility in the prephase of condensation, *Tellus*, *21*, 631-635, 1969.
- Kent, G. S., C. R. Trepte, K. M. Skeens, and D. M. Winker, LITE and SAGE II measurements of aerosols in the Southern Hemisphere upper troposphere, *J. Geophys. Res.*, *103*, 19,111-19,127, 1998.
- Klett, J. D., Stable analytic inversion solution for processing lidar returns, *Appl. Opt.*, *20*, 211-220, 1981.
- Koloutsou-Vakakis, S., C. M. Carrico, Z. Li, M. J. Rood, and J. A. Ogren, Aerosol properties and radiative forcing at an anthropogenically perturbed midlatitude Northern Hemisphere continental site, *Phys. Chem. Earth Pt C, Sol. Terr. Planet. Sci.*, *24*, 541-546, 1999.
- Koloutsou-Vakakis, S., M. J. Rood, A. Nenes, and C. Pilinis, Modeling of aerosol properties related to direct climate forcing, *J. Geophys. Res.*, *103*, 17,009-17,032, 1998.
- Müller, D., U. Wandinger, D. Althausen, I. Mattis, and A. Ansmann, Retrieval of physical properties from lidar observations of extinction and backscatter at multiple wavelengths, *Appl. Opt.*, *37*, 2260-2263, 1998.
- Reagan, J. A., M. V. Apte, T. V. Bruhns, and O. Youngbluth, lidar and balloon-borne cascade impactor measurements of aerosols: A case study, *Aerosol Sci. Technol.*, *3*, 259-275, 1984.
- Reagan, J. A., M. V. Apte, A. Ben-David, and B. M. Herman, Assessment of aerosol extinction to backscatter ratio measurements made at 694.3 nm in Tucson, Arizona, *Aerosol Sci. Technol.*, *8*, 215-226, 1988.
- Rosen, J. M., and T. Kjome, Balloon-borne measurements of the aerosol extinction-to-backscatter ratio, *J. Geophys. Res.*, *102*, 11,165-11,169, 1997.
- Rosen, J. M., R. G. Pinnick, and D. M. Garvey, Measurement of extinction-to-backscatter ratio for near-surface aerosols, *J. Geophys. Res.*, *102*, 6017-6024, 1997.
- Sasano, Y., and E. V. Browell, Light scattering characteristics of various aerosol types derived from multiple wavelength lidar observations, *Appl. Opt.*, *28*, 1670-1679, 1989.
- Seinfeld, J. H., et al., *Aerosol Radiative Forcing and Climate Change*, Nat. Res. Council, Nat. Acad. Press, Washington, D. C., 1996.
- Spinhirne, J. D., J. A. Reagan, and B. M. Herman, Vertical distribution of aerosol extinction cross section and inference of aerosol imaginary index in the troposphere by lidar technique, *J. Appl. Meteorol.*, *19*, 426-438, 1980.
- Winker, D. M., R. H. Couch, and P. McCormick, An overview of LITE: NASA's lidar in-Space Technology Experiment, *Proc. IEEE*, *84*, 164-180, 1996.
- Young, S. A., D. R. Cutten, M. J. Lynch, and J. E. Davies, Lidar-derived variations in the backscatter-to-extinction ratio in Southern Hemisphere coastal maritime aerosols, *Atmos. Environ., Part A*, *27*, 1541-1551, 1993.

T. L. Anderson, Joint Institute for the Study of Atmosphere and Oceans, Box 354235, University of Washington, Seattle, WA 98195-4235. (tadand@u.washington.edu)

ANDERSON ET AL.: AEROSOL EXTINCTION-TO-BACKSCATTER RATIO
 ANDERSON ET AL.: AEROSOL EXTINCTION-TO-BACKSCATTER RATIO
 ANDERSON ET AL.: AEROSOL EXTINCTION-TO-BACKSCATTER RATIO
 ANDERSON ET AL.: AEROSOL EXTINCTION-TO-BACKSCATTER RATIO
 ANDERSON ET AL.: AEROSOL EXTINCTION-TO-BACKSCATTER RATIO
 ANDERSON ET AL.: AEROSOL EXTINCTION-TO-BACKSCATTER RATIO
 ANDERSON ET AL.: AEROSOL EXTINCTION-TO-BACKSCATTER RATIO
 ANDERSON ET AL.: AEROSOL EXTINCTION-TO-BACKSCATTER RATIO
 ANDERSON ET AL.: AEROSOL EXTINCTION-TO-BACKSCATTER RATIO
 ANDERSON ET AL.: AEROSOL EXTINCTION-TO-BACKSCATTER RATIO

Captions

Figure 1. Laboratory test of the ability of the conventional nephelometer and the 180_i nephelometer to measure the extinction-to-backscatter ratio S for conservatively scattering latex spheres of known size and refractive index (1.59-0*i*). Uncertainty bars are based on -7% uncertainty for both scattering measurements and -2% uncertainty for particle size. Mie calculations (solid curve) use nearly monodisperse, lognormal size distributions with a geometric standard deviation of 1.03, consistent with the manufacturer's stated breadth of particle sizes.

Figure 2. Correlation plots of S versus potential controlling factors. Bondville data are shown for the two dominant air mass categories only: long Northwest (open circles) and stagnant (solid triangles). (a) S versus relative humidity. Solid curve is the continental model from Ackermann [1998]. (b) S versus σ_{ep} . Shaded dots are from an experiment on the Washington coast, separated into marine samples (squares) and continental samples (triangles). (c) S versus \dots . Shaded dots are as in Figure 2b.

Figure 1. Laboratory test of the ability of the conventional nephelometer and the 180_i nephelometer to measure the extinction-to-backscatter ratio S for conservatively scattering latex spheres of known size and refractive index (1.59- 0*i*). Uncertainty bars are based on -7% uncertainty for both scattering measurements and -2% uncertainty for particle size. Mie calculations (solid curve) use nearly monodisperse, lognormal size distributions with a geometric standard deviation of 1.03, consistent with the manufacturer's stated breadth of particle sizes.

Figure 2. Correlation plots of S versus potential controlling factors. Bondville data are shown for the two dominant air mass categories only: long Northwest (open circles) and stagnant (solid triangles). (a) S versus relative humidity. Solid curve is the continental model from Ackermann [1998]. (b) S versus σ_{ep} . Shaded dots are from an experiment on the Washington coast, separated into marine samples (squares) and continental samples (triangles). (c) S versus \dots . Shaded dots are as in Figure 2b.

Table 1. Measurements of S for Tropospheric Aerosols

Method	Location	N^a	S , sr
Slant-path lidar ^b	mixed layer, western United States (Tucson, Arizona)	81	8-75
Horizontal lidar ^c	Netherlands	10	10-50
Horizontal lidar ^d	marine surface layer, Australian coast	10	40-80
Multiwavelength lidar ^e	marine boundary layer (tropical Atlantic)	1	<30
	mixed layer over rainforest (South America)	1	43-60
	Saharan dust aloft (tropical Atlantic)	2	15-62
Backscattersonde ^f	rural, arid Southwestern United States	12	42
	rural western United States, lower troposphere	4	15-30
	rural western United States, upper troposphere	4	15-60
Spaceborne lidar ^g	smoke layers in Southern Hemisphere, upper troposphere	7	50-90
Raman lidar ^h	polluted lower troposphere, over northern Germany	2	20-40
Raman lidar ⁱ	polluted boundary layer over Leipzig, Germany	1	55-95
Raman lidar ^j	polluted lower troposphere, Indian Ocean	2	50-90
Raman lidar ^k	lower troposphere, Central United States	25	20-80
Nephelometry	northwest coastal United States ^l	3	20-70
	polluted, central United States ^m	35	30-71

^a Approximate number of independent samples.

^b Determines effective S over mixed layer at 694 nm [Spinhirne *et al.*, 1980; Reagan *et al.*, 1984, 1988].

^c A 1064-nm laser; no information on location of instrument or type of aerosol investigated [de Leeuw *et al.*, 1986].

^d A 532-nm laser aimed 2 m over ocean surface from coastal site; effect of waves not assessed [Young *et al.*, 1993].

^e S at 600 nm is constrained by Mie calculations based on the measured wavelength variation of β_p [Sasano and Browell, 1989].

^f S reported at 690 nm; method requires significant wavelength and angular adjustments, based on assumed size distributions and Mie calculations [Rosen *et al.*, 1997; Rosen and Kjome, 1997].

^g S at 532 nm is constrained to values yielding physically plausible lidar retrievals under the assumption of constant S throughout upper troposphere [Kent *et al.*, 1998].

^h S at 308 nm determined from independent extinction measurements using nitrogen-Raman; one profile [Ansmann *et al.*, 1992].

ⁱ S at 532 nm using nitrogen-Raman as above [Mller *et al.*, 1998].

^j S at 532 nm using nitrogen-Raman as above; single profile showing two pollution layers over the Maldives [Ansmann *et al.*, 2000]. The published range of S was 60-110 sr, but subsequent measurements and analysis revealed an error in the overlap correction function (Detlef Mller, personal communication, 2000). The corrected range of S is 50-90 sr as shown above.

^k S at 351 nm using nitrogen-Raman as above; profiles in lower troposphere over rural Oklahoma, eight nights [Ferrare *et al.*, 1998].

^l The method discussed herein; 532 nm; S was approximately 20 during marine flow and 60-70 during continental flow [Doherty *et al.*, 1999].

^m This work; 532 nm; assumes one independent sample every 12 hours.

Table 2a. Values and Uncertainties for Air Mass Categories (Meteorology)

	Local		Two-Day Back Trajectory		
	T_d , °C	RH, %	WS, m/s	Distance, km	Height, km
	<i>All (10 Distinct Episodes and Transitions)</i>				
Ave	10.8	54	9.8	1690	1.5
SD	4.8	20	6.1	1050	0.9
	<i>Long NW (Three Distinct Episodes)</i>				
Ave	7.7	53	13.9	2410	2.0
SD	3.2	22	5.9	1010	0.8
	<i>Stagnant (Four Distinct Episodes)</i>				
Ave	16.2	56	5.9	1020	1.0
SD	2.5	18	3.1	540	0.5

Column headings are T_d , dew point temperature; RH, relative humidity; WS, net 2-day horizontal wind speed; distance, net 2-day horizontal travel distance; and height, height of air parcel 2 days previous to sampling. Ave and SD refer to average and standard deviation.

Table 2b. Values and Uncertainties for Air Mass Categories (Aerosol Optical Properties)

	Extensive Properties			Intensive Properties		
	σ_{sp} , Mm ⁻¹	σ_{ap} , Mm ⁻¹	β_p , Mm ⁻¹ sr ⁻¹	\hat{a}	ω	S , sr
<i>All (2514 10-Min Samples)</i>						
Ave	51.7	5.3	1.06	1.9	0.864	47.4
Prec (ave)	1.2	0.3	0.09	0.2	0.006	4.7
Unc (ave)	4.3	1.2	0.14	0.6	0.021	7.2
SD_m	56.7	3.6	0.83	0.39	0.08	14.4
SD_m/N	189.0	6.0	6.9	6.5	4.0	1.9
SD_t	56.7	3.5	0.80	0.38	0.08	(12)
<i>Long NW (1230 10-Min Samples)</i>						
Ave	19.1	4.4	0.58	1.8	0.824	40.5
Prec (ave)	0.4	0.3	0.13	0.2	0.010	9.1
Unc (ave)	1.6	0.9	0.14	0.6	0.035	10.0
SD_m	10.0	3.6	0.24	0.37	0.08	12.2
SD_m/N	48.0	6.0	2.2	4.6	2.7	1.4
SD_t	10.0	3.5	0.20	0.36	0.07	(8.6)
<i>Stagnant (660 10-Min Samples)</i>						
Ave	128.0	6.7	2.09	2.2	0.946	64.3
Prec (ave)	3.2	0.4	0.18	0.2	0.003	5.7
Unc (ave)	10.7	1.9	0.27	0.6	0.014	8.5
SD_m	60.4	3.4	0.90	0.31	0.03	6.5
SD_m/N	151.0	5.7	6.4	15.5	3.0	1.3
SD_t	60.4	3.3	0.90	0.31	0.03	(4.2)

Given are aerosol optical properties at 532 nm, except ω , which refers to the wavelength dependence of scattering from 450 to 550 nm. Ave refers to average. Prec (ave) and Unc (ave) are the 95% confidence precision and total uncertainties of the average. SD_m and SD_t are the measured and estimated true standard deviations, respectively, where the latter accounts for instrumental noise, N . SD_t is highly uncertain and is thus shown in parentheses when SD_m/N is less than 2.