Hourly concentrations and light scattering cross sections for fine particle sulfate at Big Bend National Park

Susanne V. Hering[a,*], Mark R. Stolzenburg[a], Jenny L. Hand[b],
Sonia M. Kreidenweis[b], Taehyoung Lee[b], Jeffrey L. Collett Jr.[b],
David Dietrich[c], Mark Tigges[c]

[a] *Aerosol Dynamics Inc., 2329 Fourth Street, Berkeley, CA 94710, USA*
[b] *Colorado State University, Fort Collins, CO 80523, USA*
[c] *Air Resource Specialists, Inc., 1901 Sharp Point Drive, Suite E, Fort Collins, CO 80525, USA*

Abstract

Fine particle sulfate was measured continuously for 90 consecutive days, from August through October 1999, at Big Bend National Park. Measurements were made with a prototype integrated collection and vaporization cell, whereby particles are humidified and collected by impaction onto a metal strip and analyzed in place by flash-vaporization and pulsed fluorescence detection of the evolved sulfur dioxide. The time resolution was 12 min. Sulfate values are compared to 24-h integrated filter measurements. Time variation in observed concentrations are compared to hourly measurements of dry particle size distributions and to nephelometry at ambient humidity. For most of the study period the sulfate was the major constituent of the accumulation mode aerosol. The sulfate scattering cross section as a function of relative humidity is inferred by comparisons among these measurements using a two-component fine particle model wherein the nonsulfate accumulation mode volume is attributed to nonhygroscopic components.
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1. Introduction

The dependence of the scattering efficiency of ambient sulfate aerosols on relative humidity is important to assessing atmospheric visibility in our parklands (Malm et al., 2000), and to evaluating direct radiative forcing by sulfates in global climate change models. Of the chemical components identified in atmospheric aerosols, sulfate has been considered an important player in global climate because of its abundance, and its hygroscopic nature (IPCC, 2001). The humidity dependent scattering efficiency of sulfates is one of the larger uncertainties in global climate modeling (Pilinis et al., 1995; Adams et al., 2001).

The uptake of water by sulfate aerosols depends upon the chemical composition, whether acidic or neutralized, and whether found as a mixture with other constituents such as organic matter. Scattering efficiency further depends on the size distribution. Neither of these quantities is routinely measured. Several investigators have used tandem differential mobility systems to measure the uptake of water by ambient aerosols (Liu et al., 1978; McMurry and Stolzenburg, 1989; Svenningsson et al., 1994; Zhang et al., 1993; Covert and Heintzenberg, 1993; Berg et al., 1998; Cocker et al., 2001); others have combined mobility and optical analysis to extend the water uptake to larger particles sizes (Kreisberg et al., 2001). These studies have been valuable in developing visibility models, and in understanding the humidity dependence of particle scattering. Many questions remain. Most difficult is direct measurement of the dependence of ambient particle scattering on relative

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*Corresponding author. Tel.: +1-510-649-9360; fax: +1-510-649-9260.

E-mail address: susanne@aerosol.us (S.V. Hering).

humidity at high relative humidity values, above 90% RH.

Another approach to determining the effect of relative humidity on ambient particle light scattering is to measure it directly using controlled-RH nephelometry. This approach was first employed by Charlson and coworkers (Charlson et al., 1978; Covert et al., 1980; Waggoner et al., 1981), and was used to infer the composition and concentration of the sulfate-containing aerosols. If chemical composition and size distribution data are available then theoretical models of water uptake can be tested against measured relative humidity dependence of particle scattering coefficient. Malm et al. (2000) and Malm and Day (2001) used this approach with 12 or 24-h averaged chemical composition data. Their results show the relative increase in sulfate scattering over the range from 20% to 90% RH, and illustrate the differences found for two different geographical regions.

We present here a slightly different approach in which the humidity dependence of the sulfate scattering cross section is inferred from time-resolved, automated measurements. The measurements were made at Big Bend National Park, located in a remote region of southern Texas, along the Rio Grande River at the border with Mexico. The field campaign was conducted as part of the Big Bend Regional Aerosol and Visibility Observational (BRAVO) study. A prototype automated sulfate monitor provided 12-min measurement of particulate sulfate concentrations during a period of three months in the summer of 1999. The time-resolved sulfate concentrations are examined in concert with the other real-time particle measurements at the site, namely: (1) dry particle size distributions measured using a combination of electrical mobility, optical particle scattering and aerodynamic particle sizing, and (2) particle light scattering measured at ambient relative humidity using an open path nephelometer. Hourly averaged data from these instruments are evaluated to infer the dependence of the scattering cross section of sulfate aerosol on the daily fluctuations in relative humidity.

2. Experimental methods

Sulfate was measured with 12-min time resolution using an integrated collection and vaporization cell (ICVC) similar to that developed by Stolzenburg and Hering (2000) for particulate nitrate. The system, shown in Fig. 1, is enclosed in a ventilated housing to maintain ambient temperature at the point of measurement. Ambient air is sampled through a 0.25 in aluminum tube housed within a ventilated duct at a flow rate of 1 l/min. The ambient sample is drawn through an impactor precut to remove particles above 2.5 μm.
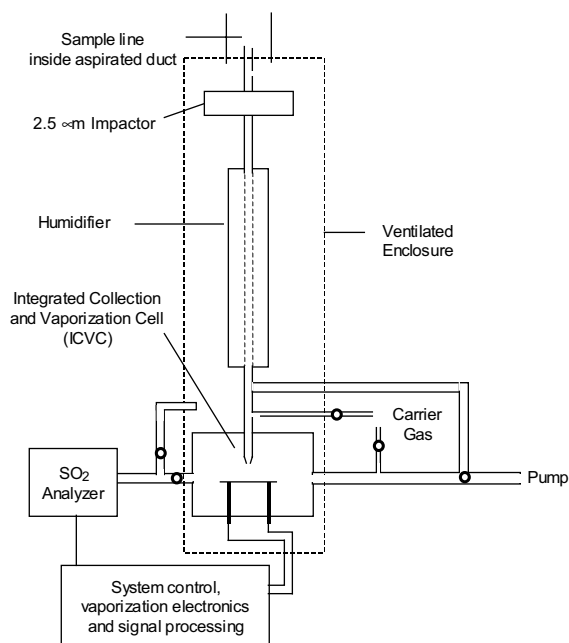

Fig. 1. Schematic of the automated sulfate system.

Smaller particles are humidified using a nafion membrane surrounded by a water sheath. The humidified particles are collected within the integrated collection and vaporization cell by impaction onto a nichrome strip. The impaction cell is operated at sonic conditions, and has a collection efficiency of 95% or greater in the size range between 0.14 and 0.8 μm.

The deposited aerosol is analyzed in place for particulate sulfur following the method of Roberts and Friedlander (1976). The nichrome strip is flash-heated in a dry air carrier gas using a 12 V battery system with an IR-shutoff. Typical heating duration is 50 ms. The sulfur dioxide concentration of the evolved vapor is quantitated using a pulsed UV-fluorescence analyzer (TEI model 43C-TL). The TEI analyzer was modified to operate at a flow rate of 1 l/min, with a pressure of 285 ± 5 Torr in the detection cell. For each analysis the baseline is read immediately before each flash, and the flash area is corrected accordingly. The system automatically cycles between a 10-min sampling period and a 2-min analysis period. Cycles automatically synchronize with the top of the hour. The system is computer controlled, providing unattended, around-the-clock operation.

The automated sulfate system was operated from 4 August to 1 November 1999 at the K-Bar site in Big Bend National Park (29°18 min N latitude, 103°10 min W longitude, elevation of 1044 m MSL). Initial setup and training was provided by Aerosol Dynamics, but thereafter, all field operations were handled by Air

Resource Specialists staff. The standard operating procedure called for the nichrome collection substrate to be changed every third day, and calibrations and field blanks to be conducted weekly.

Calibrations were done using an aqueous mixture of ammonium sulfate and oxalic acid applied directly to the collection substrate and analyzed. Standards were applied in duplicate at each of three levels, corresponding to 30, 60 and 120 ng of sulfate. Field blanks were measured by measuring the system response when sampling through a Teflon filter placed upstream of the collection cell. The SO_2 analyzer was spanned at the beginning and end of the study, using calibration gas concentrations of 50, 100 and 200 ppb SO_2. The aqueous standards showed that initially 50% of the deposited sulfate was recovered and measured as SO_2, but by the end of the three-month study the efficiency dropped to 40%. Sulfate concentrations were calculated on the basis of the aqueous standards recovery, and were corrected for the study-mean field blank of $0.15 \pm 0.10\,\mu g/m^3$.

At the same site, particle scattering was measured using an open path nephelometer (NGN-2, Optec Inc., Lowell, MI) mounted underneath a canopy to minimize solar heating. The temperature and relative humidity were recorded immediately adjacent to the nephelometer. Particle number distributions were measured under dry conditions, $15 \pm 9\%$ RH using a differential mobility analyzer (TSI 3071), optical particle counter (PMS Las-Air 1003) and aerodynamic particle sizer (TSI 3320), and converted to volume distributions as described by Hand and Kreidenweis (2002). Each day 24-h average sulfate concentrations were determined by ion chromatographic analysis of Teflon filters collected downstream of a 2.5 μm-cutpoint cyclone.

3. Quality assurance

During the setup period for the study (29 June) a set of special experiments were conducted to ascertain the independence of the system response on sample duration. Measurements were taken using sample collection periods of 150, 300, 480 and 600 s. The indicated sulfate concentration values are compared to simultaneous nephelometer readings (as size distribution measurements were not yet being made). Although the nephelometry was done at ambient relative humidity, this varied from only 41% to 51% RH, and so the light scattering coefficient was taken as a surrogate for the relative changes in $PM_{2.5}$ concentration. As shown in Fig. 2, the data for sample durations of 300–600 s fall on a common line. The respective ratios of measured sulfate concentration to light scattering coefficient, 0.30 ± 0.04, 0.31 ± 0.01 and $0.31 \pm 0.03\,g/m^2$ for the three sample durations, are not significantly different. Data at the shortest sample duration of 150 s lie above and below
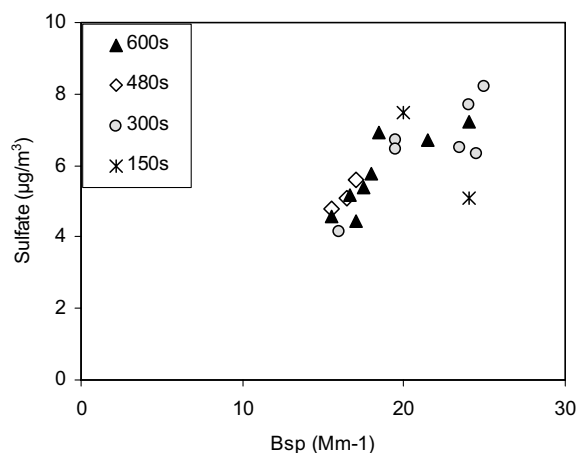

Fig. 2. Comparison of indicated sulfate concentration to particle light scattering for three different sample durations of 600, 480, 300 and 150 s. All data are from the day of 29 June 1999.

the line, indicative of the imprecision associated with the short measurement period rather than a bias.

Comparison between integrated, 24-h filter measurements, and the average of sulfate concentrations measured over the same time period by the automated system are shown Fig. 3. Results are shown separately for each of the three months of the study. For August and September, linear regression against the filter results yields $R^2 = 0.88$ and 0.90, respectively, whereas for October the correlation declined to $R^2 = 0.65$. Similarly, the response to aqueous standards decreased by 20% by the end of the study, likely due to a degradation of the flash system battery used in this prototype. While the reduced response was taken into account in the data reduction, it may also have contributed to the increased scatter. The ratio of mean sulfate concentration measured by the ICVC to that from the filter measurements is 1.10, 1.13, 1.37 for August, September and October, respectively. Linear regression over the entire study period gives $y = 0.91x + 0.76$, with $R^2 = 0.78$, where y is the automated system and x the filter value. Data are reported for more than 90% of the hours during this three-month period.

4. Study results

Fig. 4 shows 1-h concentration profiles for sulfate and sulfur dioxide at Big Bend National Park for a portion of the study. On the first few days shown, 17th through 21st August, daily sulfate maxima occurred at midnight, coincident with a maximum in the sulfur dioxide concentration. Other periods also exhibited late evening sulfate maxima, but without peaks in sulfur dioxide concentrations. Many periods showed no consistent
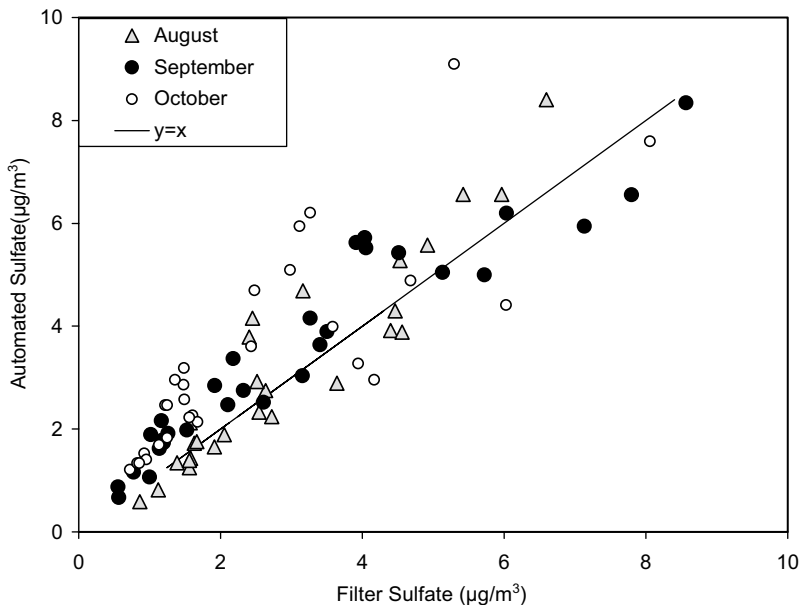
Fig. 3. Comparison of results from the automated monitor to 24-h sulfate concentrations measured by ion chromatographic analysis of PM$_{2.5}$ filters.
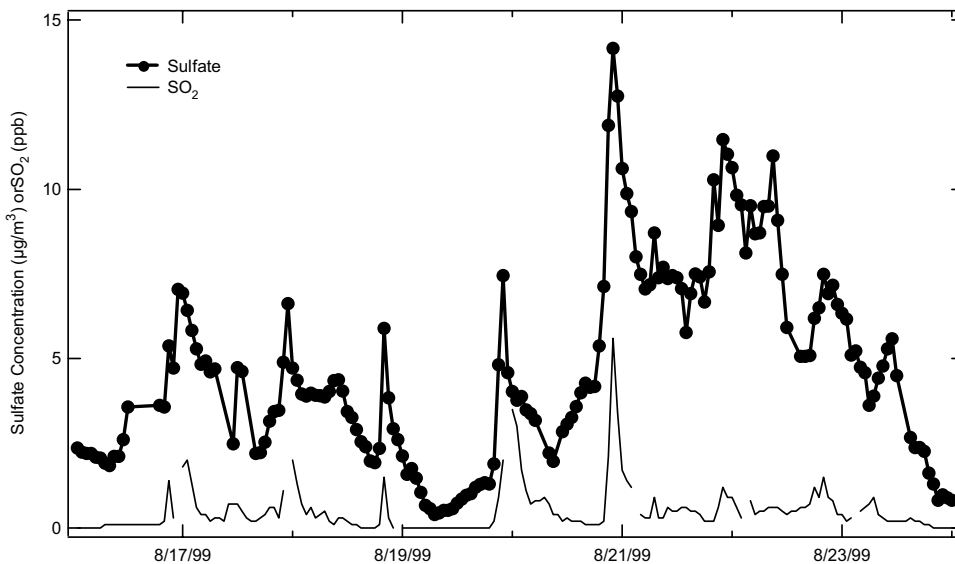

Fig. 4. Hourly sulfate and sulfur dioxide concentrations at Big Bend National Park.

diurnal pattern, with the time of maximum concentration changing from day to day. Overall, the most pronounced features are multi-day incursions of air masses with elevated sulfate, such as shown for 21–22 August.

Throughout the study, the ICVC sulfate concentrations are correlated with the accumulation mode dry volume ($R^2 = 0.89$), as reported by Hand et al. (2002). The accumulation mode is determined from the dry physical size distributions, and is defined as that portion of the spectrum below the minimum between coarse and fine mode particles when plotted as a volume distribution. The diameter that separates the accumulation and coarse modes varies from one distribution to the next, with a mean value of $0.77 \pm 0.15 \, \mu m$. The regression line of sulfate mass concentration against the accumulation mode volume has a slope of $0.79 \pm 0.01 \, g/cm^3$. For an assumed dry particle density of $1.8 \, g/cm^3$, this implies

that sulfate and its associated cations account for more than one-half of the mass. This is consistent with filter data that show, for the periods analyzed in here, that $PM_{2.5}$ sulfate and its associated cations comprised 55% of the reconstructed $PM_{2.5}$ mass. In contrast, organic aerosol, estimated as 2.1 times the measured organic carbon (Turpin and Lim, 2001), was 25% of the reconstructed fine particle mass, and was not correlated with the sulfate ($R^2 = 0.24$).

Fig. 5 shows the period from 24 August through 5 September in detail. Shown are the hourly sulfate concentration and accumulation mode volume obtained from the physical size distribution. Also shown is the total particle light scattering coefficient $B_{sp,RH}$, measured at ambient humidity, and the calculated dry particle scattering for the accumulation and coarse mode particles. Light scattering coefficients are calculated from the measured size distributions, using a refractive index inferred by matching of size data between mobility-based and optical scattering instruments in the region of overlapping data (Hand and Kreidenweis, 2002). During this period the sulfate concentration and dry accumulation mode volume are highly correlated ($R^2 = 0.96$). The coarse particle scattering is a small contribution to the total scattering. When the relative humidity is high, such as occurs at night, the nephelometer data lie distinctly higher than the calculated dry particle scattering.

The dependence of sulfate scattering on relative humidity is inferred from this data set using a two-component model for the accumulation mode aerosol. The analysis is facilitated by two factors: that sulfate is the dominant accumulation mode constituent, and that the time resolution of the sulfate concentration data is sufficient to track the daily variation in relative humidity. The accumulation mode scattering is apportioned between a sulfate constituent, assumed hygroscopic, and a nonsulfate constituent, assumed to be nonhygroscopic. The sulfate is assumed to be entirely in the accumulation mode, as indicated by the high correlation between these two parameters. For the period of analysis, integrated filter samples show that $PM_{2.5}$ nitrate comprised just 6% of the reconstructed mass and is ignored. Coarse mode particles, that is those larger than the accumulation mode, are assumed to be nonhygroscopic.

The volume fraction of nonsulfate particles in the accumulation mode, V_{NS}, was calculated by difference from the hourly accumulation mode volume. The aerosol volume associated with the sulfates was calculated from the hourly sulfate data using the study mean ammonium to sulfate molar ratio of 1.56, which corresponds to a particle density of $1.78\,g/cm^3$ and a factor 1.30 to account for its associated cations:

$$V_{NS} = V_{accm} - \left\{\frac{1.30}{1.78}\right\}SO_4^{2-}. \qquad (1)$$

The scattering attributable to sulfate and its associated water was calculated as the difference between the measured particle light scattering coefficient at ambient
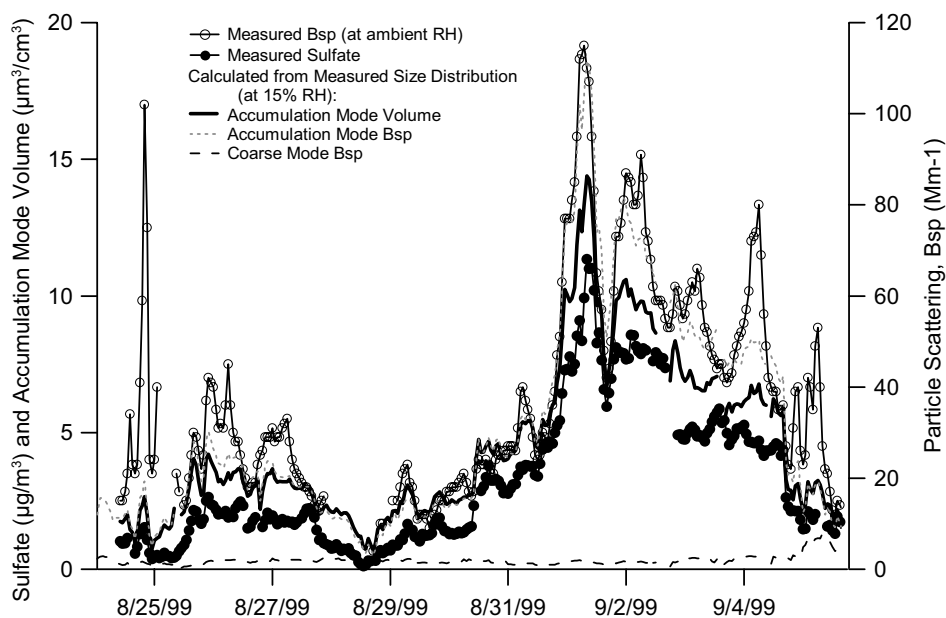

Fig. 5. Light scattering coefficient measured at ambient humidity, hourly averaged sulfate concentration measured with the ICVC system, and the dry particle accumulation mode volume, accumulation mode light scattering coefficient, and coarse mode scattering coefficient derived from physical size distribution measurements.

relative humidity, $B_{sp,RH}$, and the calculated scattering for the coarse mode and the nonhygroscopic portion of the accumulation mode, yielding an inferred relative humidity dependent scattering cross section for sulfate, $\sigma_{sulfate}$ as follows:

$$\sigma_{sulfate} = \frac{1}{SO_4^{2-}}\left\{ B_{sp,\,RH} - B_{sp,\,crs} - B_{sp,\,accm}\frac{V_{NS}}{V_{accm}}\right\}, \quad (2)$$

where $B_{sp,crs}$ and $B_{sp,accm}$ are, respectively, the coarse and accumulation mode dry particle scattering coefficients calculated from the measured size distribution and where the accumulation mode scattering for nonsulfate is assumed to be proportional to its volume fraction. While the volume apportionment of nonsulfate scattering is not exact, its contribution is small for the sulfate-dominated periods considered here. The data were screened to exclude those hourly periods for which the coarse particle scattering $B_{sp,crs}$ was more than 20% of the total calculated dry particle scattering (14% of the data set), and those periods for which sulfate did not account for at least one-half of the accumulation mode volume (another 1% of the data). All of the parameters on the right-hand side of Eq. (2) are derived from hourly averages of the automated measurements.

If the sulfate and nonsulfate materials are carried by different particles (i.e. the accumulation mode is externally mixed) and have similar size distributions and refractive indices, then Eq. (2) yields a valid approximation for the contribution of sulfate to scattering, expressed as scattering cross-section per mass, or specific scattering. For such external mixtures, $\sigma_{sulfate}$ is equivalent to the scattering efficiency of sulfate. If the sulfates are found in the same particles as nonhygroscopic components (i.e. if the accumulation mode is internally mixed), then the specific contribution of sulfate to scattering is not a strictly meaningful quantity and the calculation of sulfate's scattering efficiency requires additional information and assumptions (White, 1986). Regardless of the mixing state, if one assumes that the hygroscopic growth is entirely associated with the sulfate component, then the increase in $\sigma_{sulfate}$ with increasing RH is due solely to the increase in scattering per unit mass of sulfate resulting from the uptake of water. We refer to $\sigma_{sulfate}$ as the scattering cross section for sulfate, as this reflects the scattering from the particle attributable to its sulfate component. Although our accounting is not exact, the contribution of nonsulfate material to scattering is small for the sulfate dominated periods considered here.

Hourly values of the inferred sulfate scattering cross section from Eq. (2) are shown in Fig. 6 for the entire study period. Data are divided among the various periods with elevated sulfate. The scattering increases by a factor of 3 between 20% RH and 80% RH, and by a factor of 5 between 20% and 90% RH, in agreement with calculations by Malm et al. (2000) for ammonium

bisulfate. They are similar to the estimates of scattering as function of relative humidity for sulfates from Grand Canyon and Great Smoky Mountains National Parks. (Malm and Day, 2001) At 80% RH scattering is between 7 and $12\,m^2/g$ sulfate, consistent with scattering cross sections reported by Adams et al. (2001). For points with RH < 90% the data are fit by the equation

$$\sigma_{sulfate} = (2.02 \pm 0.10)$$
$$\left\{ 1 + \frac{(2.08 \pm 0.21)RH(0.00124 \pm 0.002)RH^2}{100 - RH}\right\},$$

where $\sigma_{sulfate}$ is in units of m^2/g of sulfate.

Only one period, between 1600 and on 24th August and 0100 25th August, had relative humidity values in excess of 90%. During this period the fraction of the accumulation mode attributed to sulfate by Eq. (1) ranged from 61% to 86%, with a mean value of 0.72. As shown in Fig. 7, the scattering cross section becomes quite large, reaching over $60\,m^2/g$ SO_4^{2-} at 97% RH. For comparison, we show the calculated scattering per unit mass of sulfate for log-normally distributed sulfuric acid and ammonium sulfate aerosols corresponding to the measured accumulation mode volume mean diameter of $0.28\,\mu m$ at 15% RH, and measured geometric standard deviation of 1.58. For ammonium sulfate the measured mean size corresponds to a dry distribution with a number mean diameter of $0.15\,\mu m$. For sulfuric acid calculations were done at a dry number mean geometric diameter of $0.12\,\mu m$ to account for the associated water at the 15% measurement relative humidity. Water uptake is taken from the hygroscopic growth data of Tang and Munkelwitz (1994). All particles were assumed to grow in equal proportion, yielding log-normal distributions with a constant geometric standard deviation and with geometric mean diameters that increase with increasing relative humidity. Calculations are done at the 550 nm wavelength and 5–175° collection geometry of the Optec nephelometer. One sees that the comparison is quite good for both ammonium sulfate and sulfuric acid. For the 24-h period that brackets these data the measured molar ratio of ammonium ion to sulfate was 1.96, one of the highest values seen in during the study. The identification of particle composition on the basis of hygroscopicity is reminiscent of the early work of Charlson et al. (1978). Importantly, these results provide experimental confirmation of calculated scattering efficiencies at high relative humidity values.

Our model attributes all hygroscopic growth to sulfate, and thus over-predicts the sulfate scattering cross section if hygroscopic organic matter is present. Dick et al. (2000) have shown that organic matter in remote areas can be hygroscopic. Their data for Great Smoky Mountains National Park indicates that the water uptake by organic matter in the range from 80%
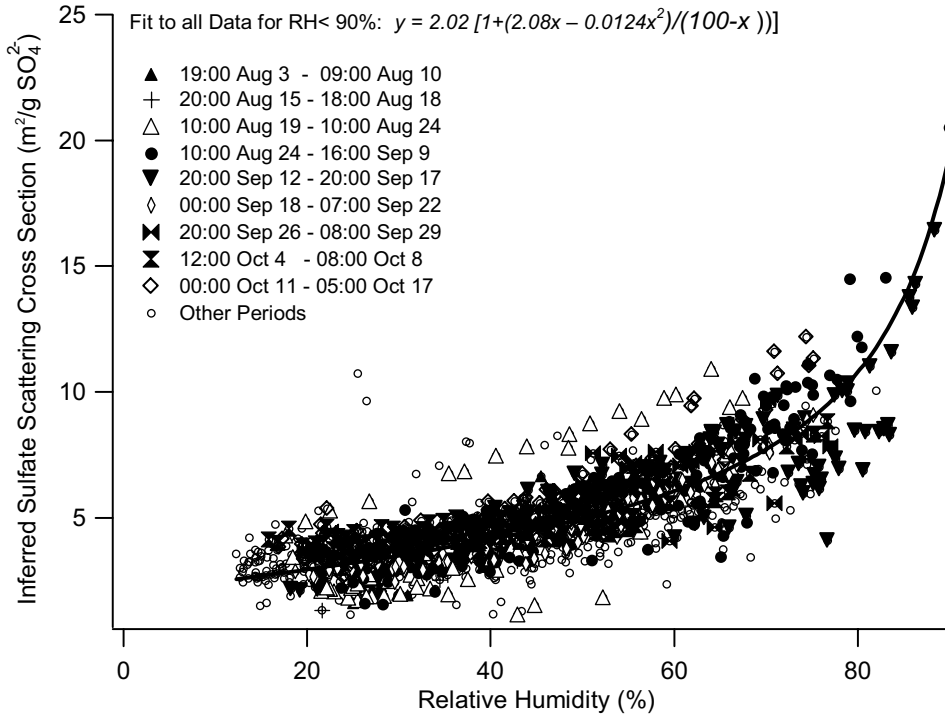
Fig. 6. Inferred light scattering cross section for sulfate aerosol as a function of relative humidity for all periods, excluding periods of dust events or low sulfate mass fraction (see text).
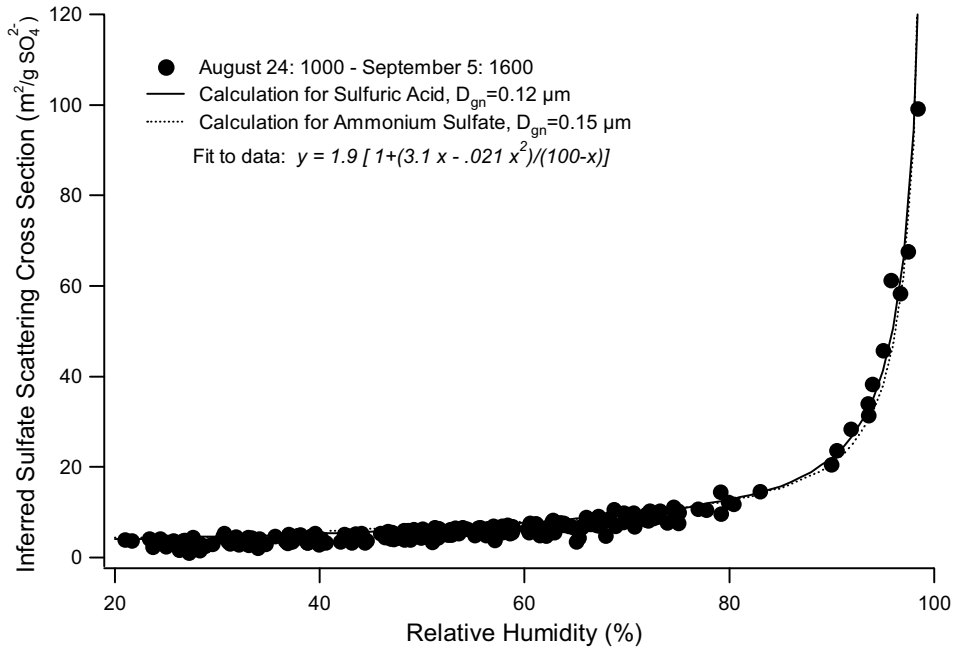
Fig. 7. Inferred light scattering cross section for sulfate aerosol as a function of relative humidity for the period from 24 August through 5 September 1999.

to 90% RH was between 25% and 50% of that for sulfate compounds. If, for the data of Fig. 7, one were to assume that the nonsulfate accumulation mode were one-half as hygroscopic as the sulfates, then the inferred cross section for sulfate in the range above 90% RH would be lower than shown by 10–30%.

To assess the importance of the hourly time resolution to this analysis, the analysis was repeated with the same data set after averaging to 24 h. For the 24-h period beginning at 0700 on 24th August, the relative humidity varied from 66% RH to 99% RH, with a mean of $86 \pm 14\%$. The sulfate light scattering cross-section calculated from the 24-h averaged data for this same time period was $34 \, m^2/g$. This lies well above the line shown in Fig. 7, which indicates a scattering cross section of approximately $17 \, m^2/g$ at 86% RH. Both values are derived from the same data set, using Eq. (2). The only difference is the averaging time for the input experimental parameters, namely nephelometer reading, dry scattering efficiencies, accumulation mode volume and sulfate concentration. The reason for the discrepancy is the nonlinear behavior of the growth curve, which is especially pronounced in the region above 80% RH. Below 70% RH the two analysis approaches yield similar results.

5. Summary

Hourly resolved data from a prototype sulfate monitor, physical size distributions at 15% RH, and light scattering at ambient RH were used to infer the relative humidity dependence of light scattering from sulfates. Data were collected at Big Bend National Park, a remote region with fine particle composition dominated by sulfates. The hourly data provided time resolution for particulate concentrations that was comparable to the daily fluctuations in relative humidity. This was used to infer the humidity dependence of the sulfate scattering assuming that other fine particles were nonhygroscopic. On average the results are comparable to theoretical results for ammonium bisulfate, as is consistent with the mean chemistry. For one day with relative humidity values in excess of 90% the aerosol behaved similarly to pure sulfuric acid or to pure ammonium sulfate. Although the simplified model presented here is applicable to sulfate-dominated airsheds only, the analysis illustrates the value of hourly concentration data in the experimental determination of the relative humidity dependence of scattering for sulfates as they are found in ambient particles. Hourly time resolution was found to be especially important for evaluating scattering at high relative humidity. Importantly, this analysis provides experimental support of theoretically derived scattering cross sections at high relative humidity values.

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