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Angular Illumination and Truncation of Three Different Integrating Nephelometers: Implications for Empirical, Size-Based Corrections

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Integrating nephelometers are widely used for monitoring and research applications related to air pollution and climate. Several commercial versions of the instrument are available and are in wide use in the community. This article reports on results from a calibration and intercomparison workshop where several units of the three most widely used nephelometer models were tested with respect to their CO_2 calibration accuracy and stability and non-idealities of their angular illumination function. Correction factors that result from the non-ideal illumination due to truncation of the sensing volumes in the near-forward and near-backward angular ranges and for non-Lambertian illumination from the light sources are presented, in particular for two models that have not previously been tested in this respect. The correction factors ranged from 0.95 to 1.15 depending on the model of nephelometer and aerosol size distribution. Recommendations for operational data analysis in context of these and previous performance tests are presented.

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1. INTRODUCTION

The integrating nephelometer provides a means of measuring light scattering coefficients of ambient aerosol particles with a high sensitivity and time resolution in a wide range of monitoring and research applications related to air pollution and climate. Accurate, direct measurements of the scattering and absorption coefficients of atmospheric aerosol particles are of importance for determining how particles affect radiative transfer and visibility in the atmosphere. The integrating nephelometer, as devised by Beuttell and Brewer (1949), measures the particulate scattering coefficient, σ_{sp}, by performing a geometrical integration of the light scattered from a sample volume illuminated by a Lambertian light source that is orthogonal to the axis of the detector. As such, the nephelometer provides a direct measure of scattering coefficient independent of the size, chemical composition, and physical state of the aerosol in an enclosed volume and it can be calibrated by gases with known scattering coefficients. Major assumptions inherent in this design are: (a) that the illumination is truly Lambertian over all angles, (b) that the light source dimension is small compared to length of the sensing volume (integration axis), and (c) that the forward and backward (near 0 and 180 degree) truncation limits of the angular integration are small. To the extent that these assumptions are not met (non-idealities in realizing the design) the measured scattering coefficient may be in error and needs to be corrected. Further description of the assumptions and non-idealities have been given by Anderson et al. (1996) and also by Moosmüller and Arnott (2003). A historical and scientific review of this type of instrument was presented by Heintzenberg and Charlson (1996).

The accuracy and uncertainty of several models of nephelometers has been determined for gas calibrations and also by closure studies using monodisperse aerosols over a range of sizes by Anderson et al. (1996) and Heintzenberg et al. (2006). These results have been evaluated by Anderson and Ogren (1998) and an empirical formula was determined to correct for the truncation and illumination non-idealities of one model of nephelometer (model 3563, TSI Inc, St. Paul, MN, USA). The effect of the non-idealities was that the measured scattering coefficient was generally less than the true value and that this systematic error is strongly dependent on particle size and more significant for particles larger than ca. 1 micrometer in diameter. Previous characterizations of nephelometers have not included direct measurement of the illumination, truncation functions for models other than the TSI 3563. Heintzenberg et al. (2006) modeled the function for two other models and estimated their illumination functions via an optimization of closure with the TSI units.

An intercomparison and calibration workshop was conducted in March of 2007 at the World Calibration Centre for Aerosol Physics (WCCAP) of the World Meteorological Organization—Global Atmospheric Watch (WMO-GAW) at the Leibniz Institute for Tropospheric Research in Leipzig, Germany, to further characterize nephelometers from three manufacturers. Our goals were: (a) to test the standard calibration and calibration procedures and (b) to test the light source's angular illumination functions and determine correction functions for the non-idealities.

2. METHODS

2.1. Instruments

Nephelometers from three manufacturers were investigated during the workshop (subsequently referred to as WCCAP2007) including the TSI model 3563, the Radiance Research model M903, and the Ecotech model 9003. Details of the manufacturer, number of units of each that were tested and their wavelength and angular integration ranges are given in Table 1. Multiple instruments were included in this study; the TSI units included manufacturing dates from 1996 to 2006 including one instrument used in the study by Anderson et al. (1996).

2.2. Light Source Characterization

The light sources were removed from the instruments and mounted on an optical bench for measurement of the angular distribution of light intensity, the illumination function. The light sources were operated identically to normal with respect to voltage and current and voltage pulse duty cycle as appropriate to the model.

A goniometer (positioning goniometer or goniometric stage) is a device used to rotate an object precisely about a fixed axis. In this case the nephelometers' light sources were fixed on an optical bench and a light collection optics on an arm fixed to the bench rotated through an angular range of 180 degrees as shown schematically in Figure 1. The angular intensity function

TABLE 1
Nephelometer models and specifications

Manufacturer	Model	Number of units	Light source	Wave-length	Bandwidth FWHM	Angular integration range
TSI St. Paul, MN USA	3563	15 3/15 for illumination	Incandescent quartz-halogen, opal glass diffuser	450, 550, 700 nm	40 nm	7° to 170°
Radiance Research Seattle, WA USA	M903	2	Xenon flash lamp, opal glass diffuser	540 nm*	40 nm	10° to 170°
Ecotech Knoxfield, VIC Australia	9003	3	7 LED tuned array, ground glass diffuser	525 nm	60 nm	12° to 165°

*The wavelength of the Radiance M903 nephelometer with a Corion CA 550S interference filter is given as 545 nm in the manufacturer's manual. The value of 540 nm in Table 1 is from a measurement of the combined effective wavelength of light source, filter, and photomultiplier detector (Anderson et al. 2003).

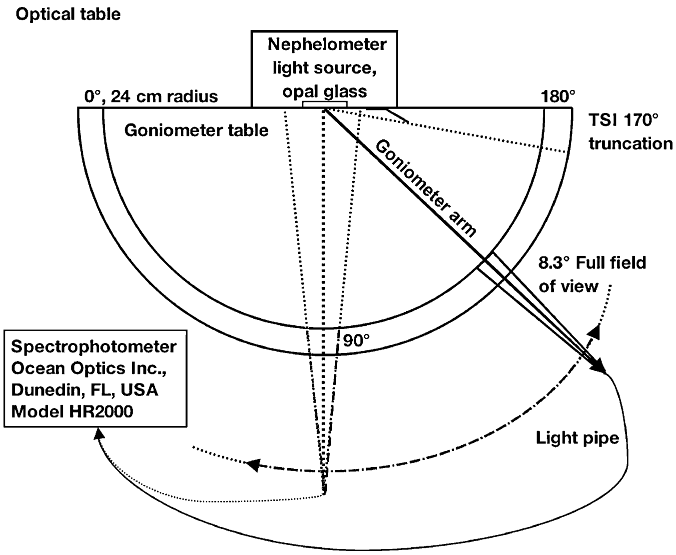

FIG. 1. Schematic of the goniometer used to measure the angular illumination function of the nephelometer light sources in WCCAP2007.

of the light sources was measured in steps of 5 degrees that were reduced to 2 degrees near 0 and 180 degrees. The goniometer arm was 24 cm in length and held a focused collection optics and light pipe connected to a spectrophotometer (model HR2000, Ocean Optics, Inc. Dunedin, FL) with a wavelength range of 400 nm to 900 nm. The full field of view of the collection optics was 8.3 degrees, which fully included the surface of the light sources in the case of the TSI and Radiance Research units, but only the most intensely illuminated, center of the Ecotech units. The illumination plane of the light source and the goniometer axis were aligned mechanically and also optically by directing a laser beam through the optical fiber (without collection lens) to the surface of the light source. The plane of motion of the arm was horizontal and perpendicular to the illumination plane of the light source. At 0 and 180 degrees the axis of the field of view of the goniometer optics was parallel to and coincident with and vertically centered on the plane of the illumination surface. The overall alignment and measurement uncertainty is estimated to be less than one degree.

2.3. Calibration Checks

Calibration checks of 15 TSI, two Radiance Research and two of the three Ecotech nephelometers were performed using measurements of filtered (i.e., particle free) air and filtered CO_2. For the TSI nephelometers, flow rates for the zero air measurements were done with the internal blower or equivalent flow at a nominal 30 l/m; CO_2 flow rate was approximately 5 l/m. The nephelometers were purged for 5 min before each measurement, followed by either a 10 min air measurement or a 5 min CO_2 measurement. The measured average values of the six scattering coefficients (450, 550, and 700 nm for total scattering and hemispheric backscattering channels) were adjusted from instrument conditions of temperature and pressure to STP conditions (standard conditions are defined here as 1013.25 mb and 273.15 K) and compared with theoretical values. Calibration checks such as these determine whether the previous instrument calibration is still appropriate or whether something has changed in the interim. For the other nephelometers, similar calibration checks were performed. For the TSI nephelometers, as received at the start of WCCAP2007 the initial calibration check results deviated from CO_2 theoretical values by –0.7% and 0.8%, average and standard deviation, respectively, with the exception of two instruments that required maintenance. The range was within $\pm 7\%$, consistent with the calibration results reported by Heintzenberg et al. (2006). Subsequent to the initial check, the instruments were all cleaned and leak checked, their lamps and photomultiplier tubes checked and replaced if necessary, and the calibrations were optimized before the intercomparisons with atmospheric aerosol. Post-optimization calibration checks on the TSI nephelometers yielded a range of average deviations from theoretical values of within $\pm 3\%$. A similar evaluation of the other models was not possible given the limited number of units.

2.4. Mie Calculations for Generic and Size Distribution Specific Corrections

We followed the methods of Anderson and Ogren (1998) and Bond et al. (2009) in which light scattering coefficients are calculated for both the Lambertian (true) and measured (neph) angular illumination functions, $\sigma_{sp,true}$ and $\sigma_{sp,neph}$ via Mie theory,

$$\sigma(x, m) = \int_0^{\pi} |S(\theta, x, m)|^2 f(\theta) d\theta, \qquad [1]$$

for size parameter $x = \pi$ d/λ, where d is particle diameter, λ is illumination wavelength, m is complex refractive index, $|S(\theta, x, m)|^2$ is scattering intensity function, and $f(\theta)$ is either the Lambertian or measured, normalized angular illumination intensity including the geometric truncation of the nephelometer collimation optics. The angular truncation limits were determined geometrically from the manufacturers stated dimensions of the mechanical components of the nephelometers. This is not a single valued limit but a range of angles over which the illumination decreases from the sine value to zero due to the finite length of the light source, cf. the sensing volume length and collimation apertures. We have used a linear approximation around these limits. In the specific case of the TSI nephelometer in the backward direction, near 180 degrees, where a shadow plate integral to the light source determines the truncation angle, the goniometer results were consistent with the geometrically determined backward truncation angle and exhibited a gradual decrease rather than a step function. For true scattering $f(\theta) = \sin(\theta)$; for nephelometer scattering, $f(\theta)$ is different as shown in Figure 2 for the several nephelometers. Equation (1) was integrated over diameter for log-normal, volume-size distributions

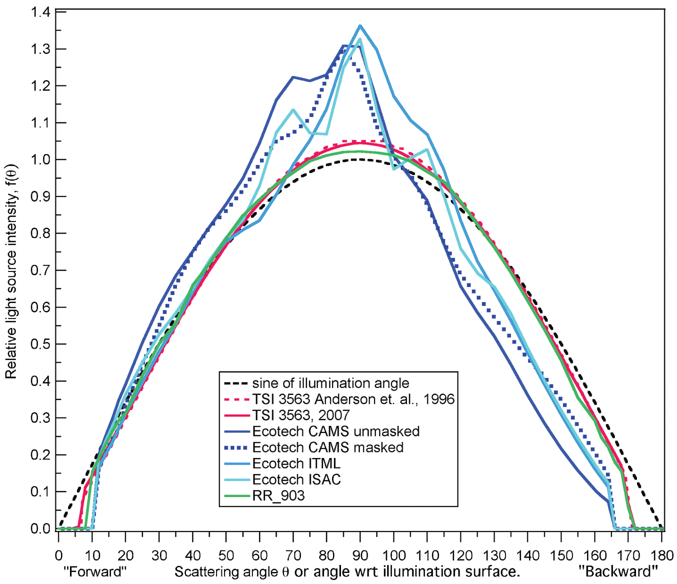

FIG. 2. Angular intensity function, $f(\theta)$, including forward and backward truncation, for the TSI 3563, Radiance Research M903 and Ecotech 9003 nephelometers at green wavelengths. For comparison purposes, each function has been normalized to the area under the reference sine curve over the truncation angle range of the TSI instrument, 7 to 170 degrees.

for a range of geometric volume-mean diameters (D_{gv}) for a geometric standard deviation, $\sigma_g = 1.8$, and refractive index, m = 1.46. These calculations were done for each of the nephelometer models and wavelengths listed in Table 1.

Subsequently, a generic and size dependent correction factor, C_{ts}, was calculated for each of the nephelometers and wavelengths for the above parameter ranges via Equation (2). Also, an Ångström exponent, $å_{sp}$, was calculated from the three wavelength data of the TSI nephelometers via Equation (3).

$$C_{ts} = \frac{\sigma_{sp,true}}{\sigma_{sp,neph}}. \quad [2]$$

$$\sigma_{sp,neph}(\lambda_1)/\sigma_{sp,neph}(\lambda_2) = (\lambda_1/\lambda_2)^{-\text{åsp}}. \quad [3]$$

3. RESULTS AND DISCUSSION

3.1. Illumination Function

The illumination intensity functions with respect to angle from the plane of the light source, $f(\theta)$, are shown in Figure 2 for the three models of nephelometer that were tested in the workshop. For comparison, the ideal, Lambertian, or sine function and the TSI light source function reported by Anderson et al. (1996) are included. The area of each of the measured functions was normalized to the sine function over the integration range 7 to 170 degrees to match the angular truncation of the TSI nephelometer for comparison. For the TSI model 3563, the WCCAP2007 result is slightly different than that of Anderson et al. (1996). The inter-instrument variability and the standard deviation of repetitive measurements on one TSI light source (not shown) were less than 1%, within the width of the plotted line. The percent deviation from ideal is within ±5% in the angular range 20 to 160 degrees but decreases to −20% close to the truncation angle limits. The illumination functions at the other TSI wavelengths, λ 450 and 700 nm, were measured and were identical to that at 550 nm in comparison to the sine function but are not shown.

The illumination function for the Radiance Research model M903 light source is relatively symmetric and close to the TSI function and the ideal sine function. Between 35 and 125 degrees the Radiance Research function has a positive deviation of about 3%; in the forward direction it is very close to sine up to the truncation limit; it deviates increasingly negatively with angle to –30% at the backward truncation angle of 170 degrees.

The illumination functions of the light sources from the three Ecotech nephelometers, shown separately, deviate from the sine function much more than those of the TSI or Radiance Research nephelometers. Due to the discrete nature of the multiple LED sources and the relatively transparent, ground-glass diffuser plate there is significant structure in the angular distribution of the light and a relatively large inter-instrument variability. The function is not symmetric and there is a significant positive deviation from sine over the angular range 15 to 100 degrees and a negative deviation over the backward scattering range. Because of the length of the Ecotech 9003 light source the entire diffuser plate could not reasonably be included in the

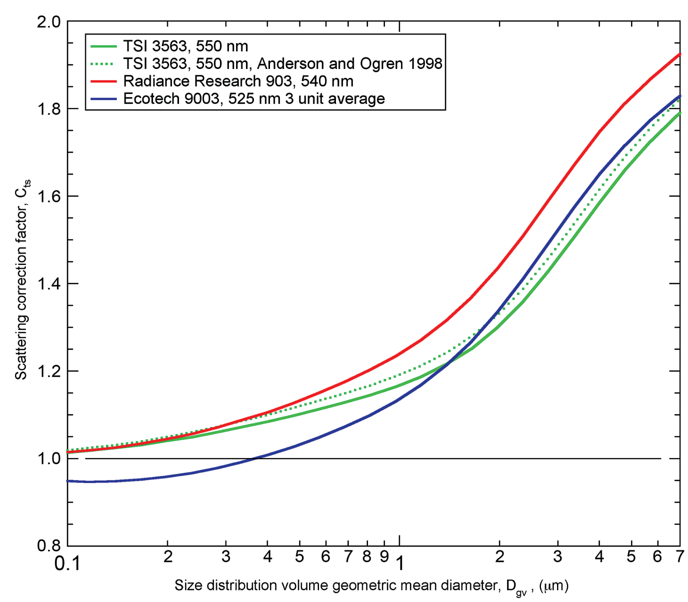

FIG. 3. Calculated generic correction factor, C_{ts}, vs. D_{gv} for three models of nephelometers accounting for truncation and angular illumination function of the light sources. The result of Anderson and Ogren (1998) from the previous illumination function for the same aerosol parameters is shown for comparison.

field of view of the goniometer but the area of most intense illumination by the LEDs was included. To test the effect of this inaccuracy, the diffuser plate was masked to the field of view of the goniometer and measured separately as shown in the figure. The resulting, masked illumination function was different but not significantly so, i.e., it was not outside the range of the unit to unit variability of the Ecotech nephelometers that were tested.

3.2. Corrections to Account for Non-Ideal Illumination and Truncation

3.2.1. Generic Correction

C_{ts} was calculated as described earlier from the measured angular illumination functions shown above and plotted vs. D_{gv} in Figure 3 with the same format as in Anderson and Ogren (1998) for green wavelengths, a log normal size distribution, $\sigma_g = 1.8$ and m = 1.46 + 0.0i and truncation limits from Table 1. This generic correction shows the small effect of the new illumination function on the correction for the TSI nephelometer and presents the previously undetermined correction factors for the Radiance Research M903 and Ecotech 9003 nephelometers.

For the TSI and Radiance Research nephelometers, the correction factors, C_{ts}, are near 1.0 at the smallest particle size, increase to 1.16 and 1.23, respectively, at a D_{gv} of 1 μm and increase monotonically to larger values in the super-micrometric range. The sense of the newly measured illumination function for the TSI nephelometers was to reduce the correction factor only minimally from 1.07 to 1.06 at a D_{gv} of 0.3 μm. The Radiance Research nephelometer has a slightly larger correction that is likely due to its larger forward truncation angle of 10° cf. the TSI truncation angle of 7°. The Ecotech nephelometer has a correction of 0.95 at D_{gv} of 0.2 μm and a negligible correction at 0.3 to 0.4 μm due to a fortuitous interaction of its truncation and non-ideal angular illumination. The illumination intensity in the forward scattering angular range from 20 to 90 degrees is greater than the ideal sine function and compensates to a large extent for the lack of intensity between 0 and 12 degrees. As with the other two instruments the correction increases at larger D_{gv} when the Ecotech nephelometer's forward truncation and insensitivity to forward scattering dominates.

3.2.2. Size, Ångström Exponent, Dependent Correction

As previous analyses and the above plots show, the correction is strongly size-dependent. Furthermore, previous analysis has shown that the Ångström exponent is also size dependent and that these two size-dependent parameters, C_{ts} and $å_{sp}$, are correlated to some extent over certain size ranges such that a self-determined correction factor can be estimated if wavelength dependent scattering is measured as in the TSI 3563 nephelometer. As in Anderson and Ogren (1998) and Bond (2009), we have plotted C_{ts} vs. $å_{sp}$ in Figure 4. For the TSI and Radiance Research nephelometers, C_{ts} ranges from 1.02 to 1.11 for a range of Ångström exponent from 2.5 to 1.0, respectively. For the Ecotech, C_{ts} ranges from 0.95 to 1.04 for the same range of Ångström exponent.

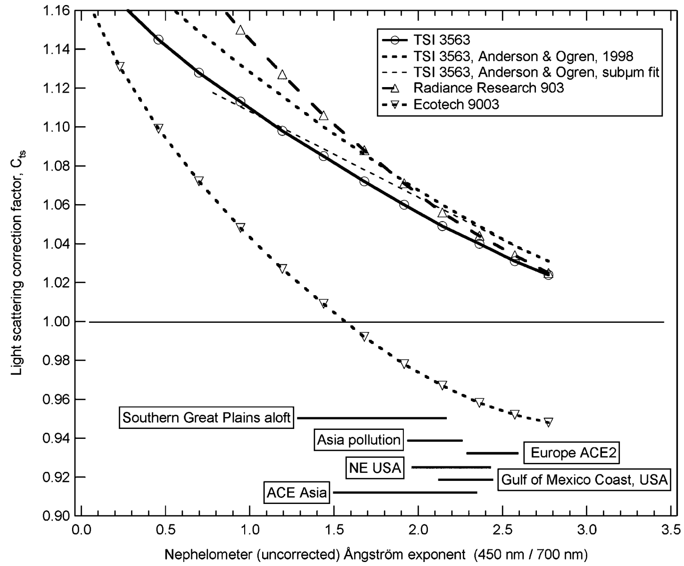

FIG. 4. Calculated generic correction factor, C_{ts}, versus Ångström exponent for the TSI 3563, Radiance Research M903 and Ecotech 9003 nephelometers based on the WCCAP2007 angular illumination function. Results based on Anderson and Ogren (1998) are shown for comparison. The horizontal bars show ranges of Ångström exponent from several regions of the globe as measured with a TSI three wavelength nephelometer (Andrews 2004; Carrico 2000; Doherty 2005; Quinn 2000; Sierau 2006).

4. CONCLUSIONS

The precision of repeated CO_2 calibration measurements at this workshop and in longer term practice among the several authors is, as stated, within 3% with a mean of 1.6% and std. dev. of 0.7%. We have no explanation for this other than to propose that it is a combination of reasonable experimental errors in the calibration and the subsequent calibration check procedures including signal noise, inconsistent purging of the sensing volume of aerosol prior to the particle free zero measurement, and of air prior to the CO_2 span measurements, errors in internal temperature values due to gradients through the nephelometer.

The correction factor resulting from the non-ideal angular truncation and illumination intensity of the TSI nephelometer remains unchanged from previous estimates within the limits of experimental error and can be estimated by the empirical, Ångström exponent-based equation of Anderson and Ogren (1998). The illumination function for the Radiance Research nephelometer is similar to that of the TSI but the correction factor is larger by a difference of 1 to 2% for submicrometric aerosol and 8 to 15% for aerosol in the coarse mode range. The Ecotech nephelometer illumination function deviates significantly from the sine function and has a correction factor of 0.95 to 1.04 depending on the size distribution of the aerosol measured. The unit to unit variability of the correction factor for the Ecotech nephelometer is of similar magnitude and without specific measurement of the particular instrument's intensity function this must be taken as an uncertainty in the measurement.

As a consequence of these results, Ecotech's Aurora model nephelometer light source has been updated to produce an angular intensity distribution function that is closer to Lambertian and more reproducible. This light source has an increased number of LEDs and a shorter axial dimension. Ecotech is currently further developing the light source to feature a diffusion plate similar to the TSI 3563.

The correction factor for the TSI instrument can be estimated by the Ångström exponent measured internally as the wavelength dependence of scattering coefficient but the correction factor for the other units must be estimated based on some knowledge of the aerosol size distribution. As explained by Bond et al. (2009) the best correction factor can be obtained by application of a measured size distribution and refractive index via Mie calculations and in the case of strongly absorbing aerosol, inclusion of the complex refractive index.

The truncation errors for these nephelometers are significant, and therefore should be accounted for when reporting aerosol light scattering coefficients. They are of similar magnitude to the calibration errors reported in section 2.3. By comparison, the error in the measured light scattering coefficient cf. the ambient or so-called dry scattering coefficient due to variation in hygroscopic growth with chemical composition of the aerosol and an uncertain or uncontrolled relative humidity in the inlet to the nephelometer and in its sensing volume can easily be much larger than the truncation, illumination error (e.g., 10% or more). The errors due to inlet losses of large particles or a poorly defined impactor- or cyclone-based size cut can also be large.

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