Comparison of Two Filter-Based Reflectance Methods to Measure the Light Absorption by Atmospheric Aerosols

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(Manuscript received 11 June 2013, in final form 13 January 2014)

ABSTRACT

Two reflectance techniques, based on Kubelka–Munk (K-M) theory and on the Beer–Lambert (B-L) law, were used to measure the absorption coefficient of aerosol particles collected on a filter. The two methods agreed, with the B-L technique being higher than the K-M method by a factor of 1.10, but with a correlation, r^2, between the two methods of 0.99. The aerosol absorption Ångström exponents (AAE) between the two methods also agreed within 0.4 and were in the range of measurements reported in the literature with other techniques. The precision of the two methods depends on the volume of air sampled, but a typical sampling scheme (100 L min^{-1}, 10 cm^2 sampling area, full day of sampling) results in a precision in the measurement of the aerosol light absorption coefficient of 0.05 Mm^{-1}.

1. Introduction

Aerosols play an important but poorly constrained role in the radiation budget of the earth–atmosphere system by scattering and absorbing solar and terrestrial radiation (direct effect; Bellouin et al. 2005) and by modulating microphysical properties of clouds (indirect effect; Rosenfeld and Lensky 1998). The radiative effects of aerosols vary greatly over space and time because they originate from multiple sources with time-varying strengths (Hinds 2012), exhibit relatively short and variable residence times against deposition (Vong et al. 2010), and include a diversity of aerosol types (Holben et al. 2001). Limitations in our understanding of the aerosol radiative effect significantly limit our ability to predict Earth's climate (Forster et al. 2007). Thus, more systematic measurements of aerosol optical properties, including the absorption coefficient, are required, in particular, with associated chemical measurements to characterize the aerosol composition and investigate aerosol processing (Arimoto et al. 2004).

In long-term monitoring programs, chemical measurements are made on aerosols concentrated on filters (Savoie et al. 2002). To integrate our measurements of the absorption coefficient into an ongoing chemical measurement program, we needed a method that could determine the optical properties from the same filters to enable close correlation of the optical and chemical parameters. This would also assist integrating the optical signature of the aerosols with models that predict aerosol transport (Hogan and Rosmond 1991). Since aerosol absorption also varies spectrally (Bergstrom et al. 2007), and this spectral variation is important in evaluating overall radiative effects (Bergstrom et al. 2003), we wanted to use a method that could allow measurement over the visible spectrum. The Interagency Monitoring of Protected Visual Environments (IMPROVE) program (Malm et al. 1994) has been making absorption measurements on filters, which are then processed for chemical species measurements, however, only at one wavelength (Campbell et al. 1995). But results from the

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DOI: 10.1175/JTECH-D-13-00131.1

IMPROVE program (Pitchford et al. 2007) show the usefulness of correlated optical and chemical measurements. The same filters we were using were also going to be used for further processing, so we also needed to minimize sample manipulation.

In summary, we wanted a simple, nondestructive technique that would enable measurement of the spectral absorption coefficient of aerosols concentrated on filters and operate in a long-term measurement program in which aerosol chemical measurements are also performed on replicate filters. There have been several workshops and method comparisons over the past 30 years on the measurement of the absorption coefficient of aerosols (Gerber and Hindman 1982b; Sheridan et al. 2005; Müller et al. 2011; Reid et al. 1998), including methods that are filter based. While earlier workshops included reflectance-based techniques (Patterson and Marshall 1982), later workshops have not included a purely reflectance-based measurement (Müller et al. 2011). A recent study, while aimed primarily at a different technique (multiangle absorption photometry; Petzold and Schönlinner 2004), showed that absorption derived from reflection alone gave good results (Petzold et al. 2005). Here, we describe a measurement technique that is a simple spectral-reflectance-based measurement and requires only one measurement of the sample filter. It is a slightly modified version of a reflectance technique used to measure the absorption of phytoplankton (Balch and Kilpatrick 1992) and is comparable to the optical reflectance method employed by Reid et al. (1998). To test this method, we compared results with a method directly derived from the classical Kubelka–Munk (K-M) theory (Kubelka and Munk 1931). In the past, measurements based on the K-M theory have been compared to other aerosol absorption techniques (Gerber and Hindman 1982a).

Any method based on measurement of aerosols concentrated on a filter will suffer from problems such as modifications of the effective aerosol shape due to collection and possible layering on the filter, relative humidity differences between measurement conditions and the environment, among other issues. In a recent review, Moosmüller et al. (2009) point out the need to compare filter-based measurements with in situ measurements such as photoacoustics and extinction-minus-scattering techniques, but then go on to point out issues with these methods. In Reid et al. (1998) a reflectance technique based on Delumyea et al. (1980) and an optical extinction technique were compared and agreed to within 17%. However, the conclusions of many of the comparison workshops point out the need for a good absolute standard to compare the various techniques (Müller et al. 2011). Because of these difficulties, it is difficult to state the absolute accuracy of the measurement technique, but based on the study by Reid et al. (1998) of a 17% agreement with extinction techniques and the study by Bond et al. (1999) where transmission measurements were found to agree within 30% with extinction techniques, a value of 30% would be a conservative estimate. Note that this is the accuracy of a single wavelength measurement. The spectral variation determined with this technique would be much more accurate, as it has been found that reflectance measurements do not require a single-scattering albedo correction factor (Petzold et al. 2005); thus, at most a simple multiplicative factor could correct all wavelengths of a measurement. Recently, Subramanian et al. (2007) found that for certain aerosols, significant modification of the aerosol occurs during filter deposition. This may introduce much more uncertainty between filter measurements and ambient measurements of absorption, and must be studied further.

2. Background

With several key assumptions (plane parallel geometry, particles small relative to layer thickness, isotropic scattering, and diffuse light field), K-M theory (Kubelka and Munk 1931; Kortüm 1969; Wendlandt and Hecht 1966) uses a two-flow approximation to radiative transfer in a medium to connect the dimensionless reflectance of an infinitely (optically) thick layer (R_∞) to the dimensionless reflectance (R_o) and transmission (T) of a more limited homogeneous layer (Kubelka and Munk 1931). The K-M parameters S and K, and the scattering and absorption coefficients commonly used in radiative transfer, can be derived from this R_o and T (Edström 2007; Mudgett and Richards 1971; Brinkworth 1972) using the following equations:

$$a = (1 - R_o^2 - T^2)/2R_o, \tag{1}$$

$$b = (a^2 - 1)^{1/2}, \tag{2}$$

$$s = \frac{1}{2db}\coth^{-1}\left[\frac{1 - aR_o}{bR_o}\right], \quad \text{and} \tag{3}$$

$$k = (a - 1)s, \tag{4}$$

where d is the geometric thickness of sample, and a and b are dimensionless parameters used by the Kubleka and Munk theory. In Eq. (3), the effective optical pathlength in one direction inside the sample of thickness d is considered to be due to the diffusely reflected light.

Since we did not have a sample with an optically thick layer of aerosol, we adapted as a comparison the method derived by Lindberg and Snyder (1973, the L-S method) based on K-M theory but designed to quantify absorption by atmospheric dust sampled on filters without an infinitely thick layer. This method requires four discrete measurements: (i) M_1—direct-diffuse reflectance of a blank filter on a black backing plate, (ii) M_2—direct-diffuse reflectance of a blank filter on a white backing plate, (iii) M_3—direct-diffuse reflectance of a sample filter on a black backing plate, and (iv) M_4—direct-diffuse reflectance of a sample filter on a white backing plate. The absolute reflectance of the white backing plate and black backing plate do not have to be perfectly 1.0 and 0.0, respectively; it is only important that there is significant contrast between the two backing plates. These four measurements can be combined to form the R_o and T needed in the K-M theory (Lindberg and Snyder 1973) as follows:

$$R_o = \frac{(M_3/M_1) - (M_4/M_2)}{M_3 - M_4 + (1/M_1) - (1/M_2)} \quad \text{and} \tag{5}$$

$$T = [(M_4 - R_o)(1/M_2 - R_o)]^{1/2}. \tag{6}$$

A variation on this approach, developed by Balch and Kilpatrick (1992), requires only three measurements: the reflectance of a sample filter ($I_{\text{samp},\lambda}$) and the reflectance of the blank filter ($I_{\text{blk},\lambda}$), each on a white background (analogous to M_2 and M_4 listed above); and dark noise ($I_{\text{dark},\lambda}$) based on the measured reflectance of a dark trap. This method is based on the assumptions that (i) the Beer–Lambert law holds for the light passing through the filter to the white backing plate and then reflects back diffusely through the sample, (ii) the filter dominates the scattering process, and (iii) scattering by particles does not significantly change the light distribution in the sample. One requirement for the Beer–Lambert law to hold is that the sample be spatially homogeneous on the filter. This is tested during measurement by making the measurement in several locations on the filter, and we have not seen any measurable inhomogeneity on our filters in the sampling area. In our application of this approach [the Beer–Lambert (B-L) method], the direct–diffuse reflectance of light normally incident on the filter was employed rather than light incident at 45° and normally reflected (Balch and Kilpatrick 1992) or light incident at 45° and reflected at 45° (Delumyea et al. 1980). This decreased any slight dependence that might be in the signal due to the light scattering phase function of the aerosols by collecting all reflected light, independent of angle. In this method the reflectance (R_λ) of aerosol sampled on a filter is given by

$$R_\lambda = \frac{I_F(\lambda)}{I_o(\lambda)} = \frac{I_{\text{samp},\lambda} - I_{\text{dark},\lambda}}{I_{\text{blk},\lambda} - I_{\text{dark},\lambda}}, \tag{7}$$

where $I_{\text{samp},\lambda}$ and $I_{\text{blk},\lambda}$ are the measured signal for the sample and blank, respectively, and are corrected for dark noise at each wavelength.

Assuming a Beer–Lambert law variation,

$$I_F(\lambda) = I_o(\lambda)e^{-x\sigma_\lambda} \quad \text{and} \tag{8}$$

$$\sigma_\lambda = -\frac{1}{x}\ln\left[\frac{I_F(\lambda)}{I_o(\lambda)}\right], \tag{9}$$

where x (m) is the effective optical pathlength through the sample and σ_λ (m^{-1}) is the spectral light absorption coefficient for the aerosol. Here x is then approximated by

$$x = n_b d. \tag{10}$$

The geometric sample thickness d (m) is assumed equal to the volume of air sampled (V) divided by the filter area (A), and n_b is an amplification factor. The effective pathlength includes both the path of the light through the sample and filter to the backing plate and the path back through the sample to where it is collected by an integrating sphere. We approximated this path as the sum of the incoming geometrical pathlength to the backing plate (d) plus the effective optical length back from the plate for totally diffuse light ($2d$) (i.e., $n_b = 3$ and, thus, $x = 3d$). Petzold et al. (2005), while comparing their multiangle absorption photometry (MAAP) technique to transmission and reflectance techniques, found a similar factor, 3.2 in their case, for samples for which the filters were lightly loaded (as were our samples). In their work they attributed the portion greater than 2 as a filter loading effect.

The absorption Ångström exponent (AAE) provides a measure of the spectral dependence of aerosol light absorption, which varies as a function of chemical composition (Russell et al. 2010). The AAE is calculated as follows:

$$\text{AAE} = -\frac{\Delta\log(\sigma_\lambda)}{\Delta\log(\lambda)}, \tag{11}$$

where $\Delta\log(\sigma_\lambda)$ and $\Delta\log(\lambda)$ are the logarithmic variation of absorption and wavelength, respectively. AAE values reported in this paper are a linear fit to the log-transformed data between 400 and 800 nm. Our data did show some slight curvature over this range, but the fitted line was within the measurement uncertainty for all points. We also report the r^2 of the line fit to the data.

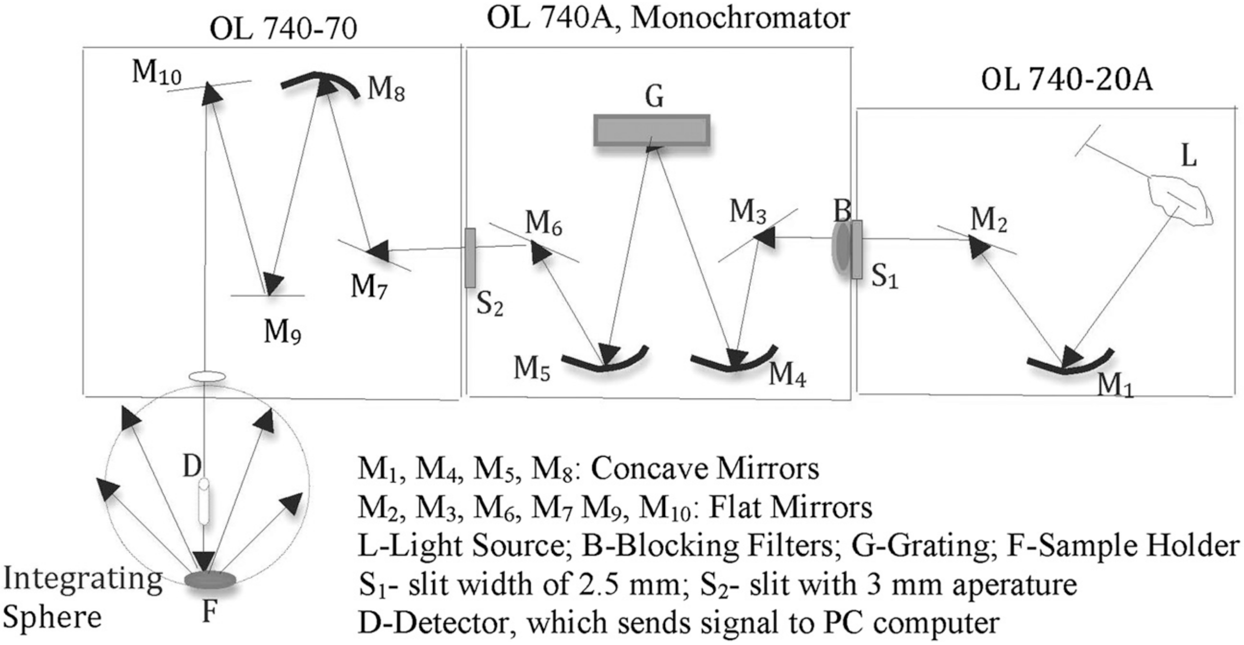

FIG. 1. An outline of the top view of filter reflectance instrumentation.

3. Experimental method

A schematic of the instrument used for the reflectance technique is shown in Fig. 1. Light from a 200-W tungsten lamp [Optronics Laboratory (OL) 740–20A] passed through a monochromator (OL 740A) and was incident on the sample. Reflected light was captured by an integrating sphere (OL 740–70) and detector. The entrance port and sample were on the sides of the integrating sphere, while the detector was perpendicular to these at the top of the integrating sphere, viewing the opposite side of the sphere. The monochromator provided wavelength selection from 350 to 1100 nm at 10-nm steps, and gave a nearly continuous spectrum from the near-ultraviolet region to the near-infrared region.

Each measurement consisted of triplet measurements (S1, S2, and S3), which are repeated until S1 and S3 agree to within the specified signal noise level of 0.5%. S1 and S3 are then averaged and this average value is saved.

Variability in aerosol loadings on filters may alter the angular distribution of reflected light and thereby bias reflectance measurements. To evaluate the potential importance of such artifacts, we measured the bidirectional reflectance distribution function (BRDF) for three samples over a range that bracketed the extremes of direct-diffuse reflectance in our data using a BRDF meter (Voss et al. 2000). No significant change (>0.03 in reflectance) was detected in the geometric shape of the BRDF between samples.

4. Results

Aerosols in near-surface, sectored (onshore) flow were sampled daily at $\sim 1.2\,\mathrm{m^3\,min^{-1}}$ with a high flow cascade impactor onto Whatman 41 filters at Miami, Florida, and Barbados by J. M. Prospero (2011, personal communication). Two samples from Miami (collected on 10–11 March and 26–27 June 2008) and one from Barbados (collected on 1–2 June 2010) were analyzed for spectral absorption with these two methods and the results were compared (Fig. 2).

All three samples exhibited similar relationships between paired data based on the two methods. A standard linear regression of the combined data for all three samples indicates that the B-L method yielded absorption coefficients that were systematically higher by a factor of 1.1 relative to those based on the L-S method when $n_b = 3$. This assumes that the incident light field stays collimated in the sample. If the light field were slightly diffuse, then it would increase this n_b factor and improve the agreement between the two methods. In fact, an n_b of 3.3 would make the agreement almost exact, and this is similar to that found in Petzold et al. (2005). However, in measurements of the BRDF of the filters, with and without dust, this value is not supported, so we are hesitant to suggest that value without other justification. The consistency of the shape of our BRDF measurements of the filters indicates that the filter itself dominates the scattering; thus, we get a nearly constant factor between the two methods, independent of the absorption, over the range sampled.

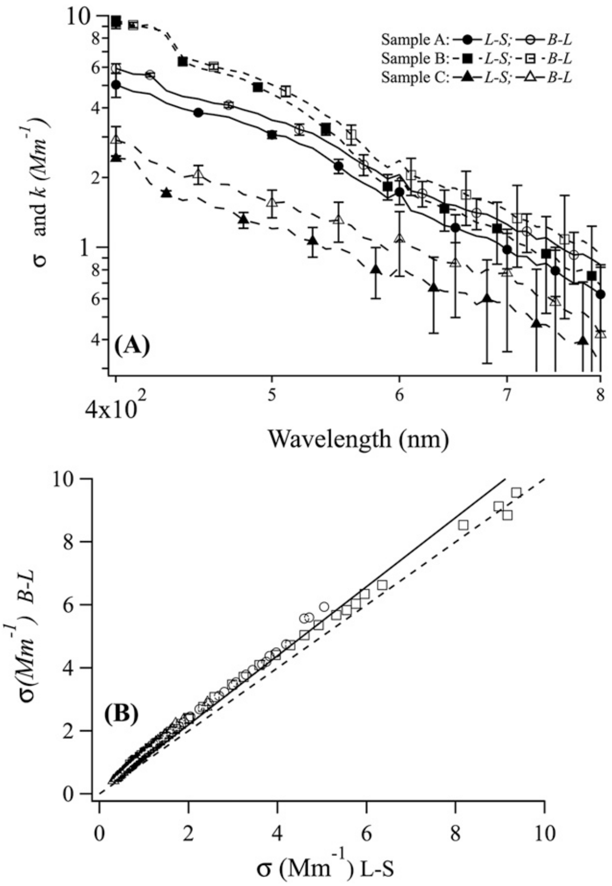

FIG. 2. (a) Spectral absorption of aerosols obtained from direct–diffuse reflectance data using B-L and L-S model techniques. (b) Comparison of the two methods; the line fit between the two methods gives a slope of 1.10 ± 0.01 ($r^2 = 0.997$). Symbols have the same shapes as in (a). The dashed line represents the 1:1 line. Error bars shown are for the standard deviation of five measurements of each sample.

There are three major sources of uncertainty in the method. The first is the uncertainty associated with the measurement itself. Each reflectance measurement has an uncertainty of approximately 0.5%. To this one must add the uncertainty due to the dark reading to get reflectance in Eq. (1). Our signal was lower in the blue due to our light source (200-W tungsten lamp). Thus, for wavelengths < 450 nm the dark reading was significant and added to the variability, but for wavelengths > 450 nm it was generally negligible. There are many more calculations and measurements in the L-S method than the B-L method, which could increase the noise in this method. However, in our measurements, we did not see an appreciable difference between the two techniques in the standard deviation of the replicate measurements.

The second source of uncertainty is due to inhomogeneity of sample deposition on the filter. Each measurement area is approximately 3 cm^2 of the typically 10–50 cm^2 available area onto which the aerosol has been deposited. During our field study, we make measurements of three areas of each filter and rotate the filter each time. In this study we made five independent measurements of each sample and found that the inhomogeneity was insignificant. Note that the standard deviation between measurements for sample B increased toward the red wavelengths. Since the absorption is also decreasing at these wavelengths, the percent standard deviation was large, rising to approximately 70% for this sample.

In practice, the measurement provides $\sigma n_b d$ for the filters, and d is derived from the volume of air sampled through an area on the filter. We found that the standard deviation, as expressed by $\sigma n_b d$, was a constant 0.007 throughout the wavelength range and for all three samples, only rising slightly near 400 nm where the lamp source was weaker. When this uncertainty is expressed as a percent of the absorption, it then varies, dependent on the value of the absorption coefficient and d. If the filter sampling area is 10 cm^2, and 100 L min^{-1} are drawn through the filter, then d is 10^{-4} Mm min^{-1}. With an uncertainty of 0.007 for the measurement of the time-integrated sample, this uncertainty in the σ measurement would be 70 min Mm^{-1}. For a full sampling day (1440 min) this uncertainty is then 0.05 Mm^{-1}.

The above-mentioned sources of uncertainty are generally random in nature and thereby influence precision but typically do not cause a systematic bias. The last source of uncertainty relates to the assumptions and associated approximations required in each technique and accounts for the systematic difference evident in Fig. 2. This is also probably the largest source of error. Unambiguous characterization would require a comprehensive intercomparison of methods, including analysis of standard reference materials, which still presents difficulties (Müller et al. 2011).

The spectral shape is consistent between the two methods, resulting in similar values for the AAE. Sample B had the strongest spectral variation, with an AAE of 3.93 ± 0.05, $r^2 = 0.992$ (L-S); and 3.49 ± 0.05, $r^2 = 0.992$ (B-L). Sample A was close to this, with an AAE of 3.14 ± 0.05, $r^2 = 0.991$ (L-S); and 2.90 ± 0.04, $r^2 = 0.994$ (B-L). Sample A agreed very well with the value estimated by Russell et al. (2010) for mineral dust of around 3, but sample B had even stronger spectral dependence. Moosmüller et al. (2012) have shown the AAE, based on measurements at 405 and 870 nm, for various samples of mineral dust, ranged from 2.5 to 3.9. However, Marley et al. (2010) have shown that the calculation of the AAE can depend on the number of wavelengths sampled. The analysis of replicate filters for samples A and B revealed a dust concentration of 42 μg m^{-3} for sample A and

$63\,\mu\mathrm{g\,m^{-3}}$ for sample B. The AAE for sample C was lower (2.60 ± 0.05, $r^2 = 0.988$, and 2.47 ± 0.04, $r^2 = 0.989$, for the L-S and B-L methods, respectively), which put this sample between the numbers suggested by Russell et al. (2010) for biomass burning (2.0) and mineral dust (3.0). The concentration of mineral dust on this filter was much less, $1.8\,\mu\mathrm{g\,m^{-3}}$, which is also reflected in the much lower absolute values of absorption. However, the concentration of $\mathrm{nssSO_4^{2-}}$ was twice as high as in sample B ($4.3\,\mu\mathrm{g\,m^{-3}}$), indicating a possible pollution influence ($\mathrm{nssSO_4^{2-}}$ was not reported for sample A).

Another factor in the use of the B-L method is the sensitivity or detection limit for the aerosol absorption coefficient. If the minimum detectable difference in reflectance between the blank filter and aerosol loaded filter is 0.5%, with a typical flow values of $100\ \mathrm{L\,min^{-1}}$ and a filter area of $20\,\mathrm{cm^2}$, then the detection limit is $2.3 \times 10^{-2}\,\mathrm{Mm^{-1}}$ for a 24-h sampling period. As the sampling period decreases, this detection limit will increase, and is $3.3 \times 10^1\ \mathrm{min\ Mm^{-1}}$.

5. Conclusions

While these two techniques qualitatively agree quite well, Beer–Lambert law seems to overestimate the absorption data by a factor of approximately 1.10. The slightly higher values of spectral absorption data with the B-L technique than the Lindberg–Snyder technique might be due to our assumption that the optical pathlength while traversing the sample to the backing filter is the geometric thickness of sample. It has been reported that the incident light field may quickly become diffuse within the sample due to multiple scattering (Kortüm 1969), thus our value of $n_b = 3$ maybe slightly low.

The AAE calculated with the data agreed well between the two methods. With both methods, since the absorption data are obtained from replicate filters for which chemical analysis was done, the correlation between the optical and chemical datasets will be straightforward with this technique.

The B-L method is simple and requires only one measurement of the sample, and thus it reduces the handling of the filter (several replicates are recommended) and provides a useful estimate of the aerosol absorption. As a filter measurement, it is easy to correlate with chemical measurements made on the same filter or replicate filters. There are issues with the representativeness of the aerosol once deposited on the filter, particularly in light of very recent work (Subramanian et al. 2007), so there needs to be further study of this issue and in the development of an absolute standard (Müller et al. 2011). Additionally, given that every technique presently available has limitations, further work must be done on more accurate techniques to measure this important parameter.

Acknowledgments. This work was supported by the Atmospheric Chemistry Division of NSF under Grant AGS-0541566. We also thank Dr. Joseph Prospero for supplying the aerosol filters that were used in this study.

REFERENCES

Arimoto, R., and Coauthors, 2004: Chemical composition of atmospheric aerosols from Zhenbeitai, China, and Gosan, South Korea, during ACE-Asia. *J. Geophys. Res.,* **109,** D19S04, doi:10.1029/2003JD004323.

Balch, W. M., and K. A. Kilpatrick, 1992: Particulate reflectance measurements of phytoplankton. *J. Plankton Res.,* **14,** 721–735, doi:10.1093/plankt/14.5.721.

Bellouin, N., O. Boucher, J. Haywood, and M. S. Reddy, 2005: Global estimate of aerosol direct radiative forcing from satellite measurements. *Nature,* **438,** 1138–1141, doi:10.1038/nature04348.

Bergstrom, R. W., P. Pilewskie, B. Schmid, and P. B. Russell, 2003: Estimates of the spectral aerosol single scattering albedo and aerosol radiative effects during SAFARI 2000. *J. Geophys. Res.,* **108,** 8474, doi:10.1029/2002JD002435.

——, ——, P. B. Russell, J. Redemann, T. C. Bond, P. K. Quinn, and B. Sierau, 2007: Spectral absorption properties of atmospheric aerosols. *Atmos. Chem. Phys.,* **7,** 5937–5943, doi:10.5194/acp-7-5937-2007.

Bond, T. C., T. L. Anderson, and D. Campbell, 1999: Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols. *Aerosol Sci. Technol.,* **30,** 582–600, doi:10.1080/027868299304435.

Brinkworth, B. J., 1972: Interpretation of the Kubelka–Munk coefficients in reflection theory. *Appl. Opt.,* **11,** 1434–1435, doi:10.1364/AO.11.001434.

Campbell, D., S. Copeland, and T. Cahill, 1995: Measurement of aerosol absorption coefficient from Teflon filters using integrating plate and integrating sphere techniques. *Aerosol Sci. Technol.,* **22,** 287–292, doi:10.1080/02786829408959747.

Delumyea, R. G., L. C. Chu, and E. S. Macias, 1980: Determination of elemental carbon component of soot in ambient aerosol samples. *Atmos. Environ.,* **14,** 647–652, doi:10.1016/0004-6981(80)90047-5.

Edström, P., 2007: Examination of the revised Kubelka–Munk theory: Consideration of modeling strategies. *J. Opt. Soc. Amer.,* **24A,** 548–556, doi:10.1364/JOSAA.24.000548.

Forster, P., and Coauthors, 2007: Changes in atmospheric constituents and in radiative forcing. *Climate Change 2007: The Physical Science Basis,* S. Solomon, Eds., Cambridge University Press, 129–234.

Gerber, H. E., and E. E. Hindman, 1982a: Data summary. *Light Absorption by Aerosol Particles,* H. Gerber and E. Hindman, Eds., Spectrum Press, 379–393.

——, and ——, 1982b: Light absorption by aerosol particles: First International Workshop. *Appl. Opt.,* **21,** 370, doi:10.1364/AO.21.000370.

Hinds, W. C., 2012: *Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles.* John Wiley and Sons, 483 pp.

Hogan, T. F., and T. E. Rosmond, 1991: The description of the Navy Operational global Atmospheric Prediction System's spectral forecast model. *Mon. Wea. Rev.,* **119,** 1786–1815, doi:10.1175/1520-0493(1991)119<1786:TDOTNO>2.0.CO;2.

Holben, B. N., and Coauthors, 2001: An emerging ground-based aerosol climatology: Aerosol optical depth from AERONET. *J. Geophys. Res.,* **106,** 12 067–12 097, doi:10.1029/2001JD900014.

Kortüm, G., 1969: *Reflectance Spectroscopy*. Springer-Verlag. 376 pp.

Kubelka, P., and F. Munk, 1931: Ein Beitrag zur Optik der Farbanstriche. *Z. Tech. Phys.,* **12,** 593–601.

Lindberg, J. D., and D. G. Snyder, 1973: Determination of the optical absorption coefficient of powdered materials whose particle size distribution and refractive indices are unknown. *Appl. Opt.,* **12,** 573–578, doi:10.1364/AO.12.000573.

Malm, W. C., J. F. Sisler, D. Huffman, R. A. Eldred, and T. A. Cahill, 1994: Spatial and seasonal trends in particle concentration and optical extinction in the United States. *J. Geophys. Res.,* **99,** 1347–1370, doi:10.1029/93JD02916.

Marley, N. A., J. S. Gaffney, V. Rajaram, and E. V. Fischer, 2010: Determining aerosol Ångström absorption coefficients: Comparison of full spectrum integrating sphere reflection spectroscopy with 3 and 7 wavelength filter absorption methods. *12th Conf. on Atmospheric Chemistry/Second Symp. on Aerosol–Cloud–Climate Interactions,* Atlanta, GA, Amer. Meteor. Soc., Paper J17.5. [Available online at https://ams.confex.com/ams/90annual/techprogram/paper_165257.htm.]

Moosmüller, H., R. K. Chakrabarty, and W. P. Arnott, 2009: Aerosol light absorption and its measurement: A review. *J. Quant. Spectrosc. Radiat. Transfer,* **110,** 844–878, doi:10.1016/j.jqsrt.2009.02.035.

——, J. P. Engelbrecht, M. Skiba, G. Frey, R. K. Chakrabarty, and W. P. Arnott, 2012: Single scattering albedo of fine mineral dust aerosols controlled by iron concentration. *J. Geophys. Res.,* **117,** D11210, doi:10.1029/2011JD016909.

Mudgett, P. S., and L. W. Richards, 1971: Multiple scattering calculations for technology. *Appl. Opt.,* **10,** 1485–1502, doi:10.1364/AO.10.001485.

Müller, T., and Coauthors, 2011: Characterization and intercomparison of aerosol absorption photometers: Result of two intercomparison workshops. *Atmos. Meas. Tech.,* **4,** 245–268, doi:10.5194/amt-4-245-2011.

Patterson, E. M., and B. T. Marshall, 1982: Diffuse reflectance and diffuse transmission measurements of aerosol absorption at the First International Workshop on light absorption by aerosol particles. *Appl. Opt.,* **21,** 387–393, doi:10.1364/AO.21.000387.

Petzold, A., and M. Schönlinner, 2004: Multi-angle absorption photometry—A new method for the measurement of aerosol light absorption and atmospheric black carbon. *Aerosol Sci.,* **35,** 421–441, doi:10.1016/j.jaerosci.2003.09.005.

——, H. Schloesser, P. J. Sheridan, W. P. Arnott, J. A. Ogren, and A. Virkkula, 2005: Evaluation of multiangle absorption photometery for measuring aerosol light absorption. *Aerosol Sci. Technol.,* **39,** 40–51, doi:10.1080/027868290901945.

Pitchford, M., W. Malm, B. Schichtel, N. Kumar, D. Lowenthal, and J. Hand, 2007: Revised algorithm for estimating light extinction from IMPROVE particle speciation data. *J. Air Waste Manage. Assoc.,* **57,** 1326–1336, doi:10.3155/1047-3289.57.11.1326.

Reid, J. S., P. V. Hobbs, C. Liousse, J. V. Martins, R. E. Weiss, and T. F. Eck, 1998: Comparisons of techniques for measuring shortwave absorption and black carbon content of aerosols from biomass burning in Brazil. *J. Geophys. Res.,* **103,** 32 031–32 040, doi:10.1029/98JD00773.

Rosenfeld, D., and I. M. Lensky, 1998: Satellite-based insights into precipitation formation processes in continental and maritime convective clouds. *Bull. Amer. Meteor. Soc.,* **79,** 2457–2476, doi:10.1175/1520-0477(1998)079<2457:SBIIPF>2.0.CO;2.

Russell, P. B., and Coauthors, 2010: Absorption Ångström exponent in AERONET and related data as an indicator of aerosol composition. *Atmos. Chem. Phys.,* **10,** 1155–1169, doi:10.5194/acp-10-1155-2010.

Savoie, D. L., R. Arimoto, W. C. Keene, J. M. Prospero, R. A. Duce, and J. N. Galloway, 2002: Marine biogenic and anthropogenic contributions to non-sea-salt sulfate in the marine boundary layer over the North Atlantic Ocean. *J. Geophys. Res.,* **107,** 4356, doi:10.1029/2001JD000970.

Sheridan, P. J., and Coauthors, 2005: The Reno Aerosol Optics Study: Overview and summary of results. *Aerosol Sci. Technol.,* **39,** 1–16, doi:10.1080/027868290901891.

Subramanian, R., C. A. Roden, P. Boparai, and T. C. Bond, 2007: Yellow beads and missing particles: Trouble ahead for filter-based absorption measurements. *Aerosol Sci. Technol.,* **41,** 630–637, doi:10.1080/02786820701344589.

Vong, R. J., I. J. Vong, D. Vickers, and D. S. Covert, 2010: Size-dependent aerosol deposition velocities during BEARPEX'07. *Atmos. Chem. Phys.,* **10,** 5749–5758, doi:10.5194/acp-10-5749-2010.

Voss, K. J., A. Chapin, M. Monti, and H. Zhang, 2000: An instrument to measure the bi-directional reflectance distribution function (BRDF) of surfaces. *Appl. Opt.,* **39,** 6197–6206, doi:10.1364/AO.39.006197.

Wendlandt, W. W., and H. G. Hecht, 1966: *Reflectance Spectroscopy*. Wiley-Interscience, 298 pp.