In Situ Measurement of Aerosol Light Absorption and Single-Scattering Albedo at the NSA and SGP CART Sites

P. J. Sheridan, [1]D. J. Delene, and J. A. Ogren
National Oceanic and Atmospheric Administration
Climate Monitoring and Diagnostics Laboratory
Boulder, Colorado

[1]Also with Cooperative Institute for Research in Environmental Sciences
University of Colorado
Boulder, Colorado

Introduction

The National Oceanic and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory (CMDL) maintains four surface aerosol-monitoring stations in North America. The reasons for conducting aerosol measurements at these stations are (1) to characterize means, variabilities, and trends of climate-forcing properties of different types of aerosols, and (2) to understand the factors that control these properties. Two of these stations, at Bondville, Illinois, and at Lamont, Oklahoma, are rural mid-continental sites. The other two stations, at Barrow, Alaska, and at Sable Island, Nova Scotia, are remote coastal sites. The Lamont station is operated in collaboration with the Atmospheric Radiation Measurement (ARM) Program at the Southern Great Plains (SGP) Cloud and Radiation Testbed (CART) site, while the Barrow station is operated in collaboration with ARM at the North

Figure 1. NOAA CMDL North American Aerosol Stations. The SGP and NSA Stations are operated in collaboration with the ARM Program.

Slope of Alaska (NSA) CART site. In this paper, we describe measurements of the aerosol light absorption coefficient (σ_{ap}), the aerosol light scattering coefficient (σ_{sp}), and the aerosol single-scattering albedo (ω_0, $\omega_0 = \sigma_{sp}/[\sigma_{sp} + \sigma_{ap}]$) at the four North American sites.

Aerosol Measurements

Aerosol particles are sampled at all four of the NOAA regional aerosol stations through 10-m-high stacks. Relative humidity (RH) is maintained below 40% by gentle heating of the air stream if necessary. An inertial impactor removes all particles larger than ~10 μm aerodynamic diameter. A second downstream impactor, through which the aerosols are passed every other 6-min cycle, periodically removes all particles larger than ~1 μm aerodynamic diameter. In this way, both submicrometer and total (sub-10 μm) aerosol fractions are identified.

The primary aerosol measurements made at the aerosol stations are the σ_{sp} and the σ_{ap}. All stations are equipped with integrating nephelometers (TSI, Inc. Model 3563) that measure the σ_{sp} over two ranges of angular integration (7° to 170° and 90° to 170°) and at three visible wavelengths (450, 550, and 700 nm). Corrections were performed that adjust the σ_{sp} values for angular truncation errors (Anderson and Ogren 1998) and also correct to conditions of standard temperature and pressure (STP, 1013.25 mbar and 0°C), which is what the absorption measurements are referenced to. The primary measurements are used to calculate the hemispheric backscatter fraction (*b*), the Angstrom exponents (*a*), and ω_0. Ancillary measurements include condensation nucleus (CN) concentration (all stations, aerosol size distributions (at the SGP site), and aerosol hygroscopic growth factor *(f[RH]), at the* SGP site).

Of particular interest in this study is the measurement of σ_{ap}. We use a light absorption photometer (Radiance Research Model PSAP) that continuously calculates σ_{ap} at 565 nm through a differential filter-based measurement. The principle of operation is to measure the change in light transmission

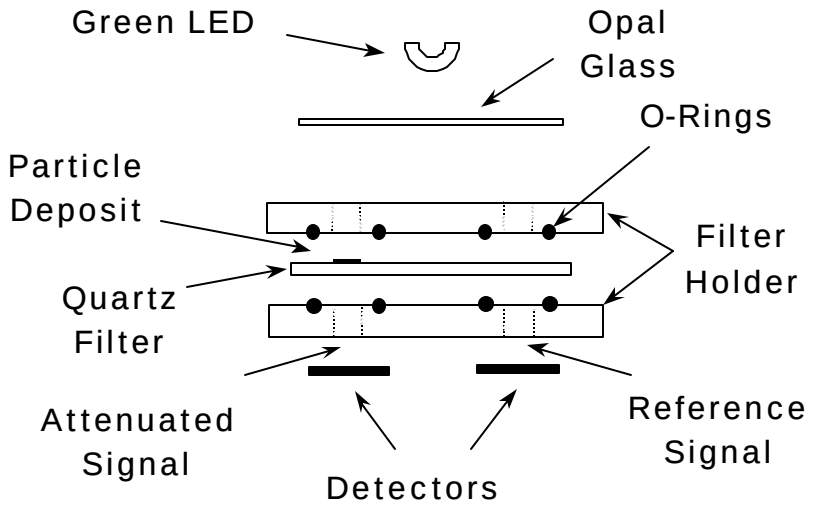

Figure 2. Schematic of the PSAP instrument.

through a filter on which particles are collected and compare that with the change in transmission through a filter section where no particles are depositing. Figure 2 shows a schematic of the PSAP.

The PSAP is calibrated using the methods described in Bond et al. (1999). The calibration uses a reference absorption determined as the difference between light extinction (measured with an optical extinction cell) and light scattering (measured using a TSI nephelometer) by suspended particles. The calibration utilizes spherical particles formed by nebulizing water solutions ammonium sulfate, nigrosin, and various mixtures of the two. Single-scattering albedos of the nebulized particles were in the range of 0.5 to 1.0. The two major findings of this calibration study were that the PSAP (1) exhibits a significant response to nonabsorbing particles and (2) overestimates absorption at 550 nm (adjusted with this calibration from 565 nm) due to suspended particles by typically ~25% to 30%. A correction scheme was developed that corrects the PSAP reported absorption value such that

$$\sigma_{ap} = (\sigma_{adj} - K_1\sigma_{sp}) / K_2 \quad (1)$$

where σ_{ap} is the true atmospheric aerosol light absorption coefficient at 550 nm, σ_{adj} is the instrument-reported absorption coefficient at 565 nm after flow and spot size corrections have been applied, and σ_{sp} is the aerosol light scattering coefficient (uncorrected for truncation errors) at 550 nm measured by a TSI nephelometer. The PSAP response to scattering was determined to be ~2% ($K_1 = 0.02$). Figure 3 shows the relationship between the reference absorption (Bond et al. 1999) and the PSAP absorption measurement. The PSAP response to absorption was found to be ~22% too high ($K_2 = 1.22$).

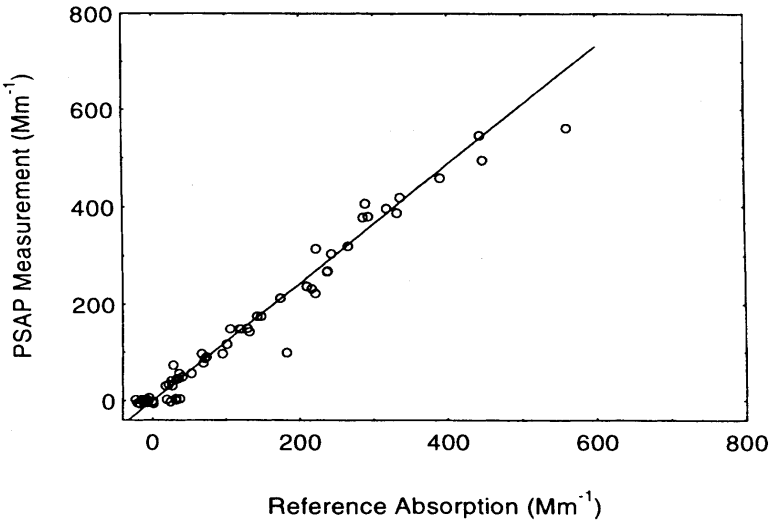

Figure 3. PSAP measurement with 2% of measured scattering subtracted versus reference absorption. The slope of the line is 1.22 (from Bond et al. [1999]).

Measurement uncertainties for filter-based light absorption measurements are unfortunately rather large. Flow rate and spot size uncertainties are each ~2%. Instrument precision (unit-to-unit variability) is around 6%. Uncertainties associated with the calibration constants K_1 and K_2 are ~100% and 9%,

respectively. Instrument noise contributes another few percent to the overall uncertainty. Assuming a 60-second averaging time and aerosol concentrations typical of the SGP site, the total estimated uncertainty of the PSAP light absorption measurement is ~30%.

Results and Discussion

Figure 4 shows annual cycles of hourly-average σ_{ap} and ω_0 data for the four CMDL North American aerosol-monitoring stations. The 12 monthly distributions are shown, along with an annual average distribution to the far right of each plot. The ends of each box, the ends of the extension lines, and the line across each box represent the 25th and 75th percentiles, the 5th and 95th percentiles, and the median of each distribution, respectively. Bondville, Illinois, (BND) typically shows the highest σ_{ap} values, followed by SGP, WSA, and NSA, although there is significant month-to-month variability. An increase in σ_{ap} is observed at the mid-continental sites SGP and BND during the autumn. The ω_0 shows a decrease at this time that is directly related to the increase in light absorption. These autumn changes in σ_{ap} and ω_0 may be related to regional-scale agricultural (e.g., crop harvesting or field burning) or

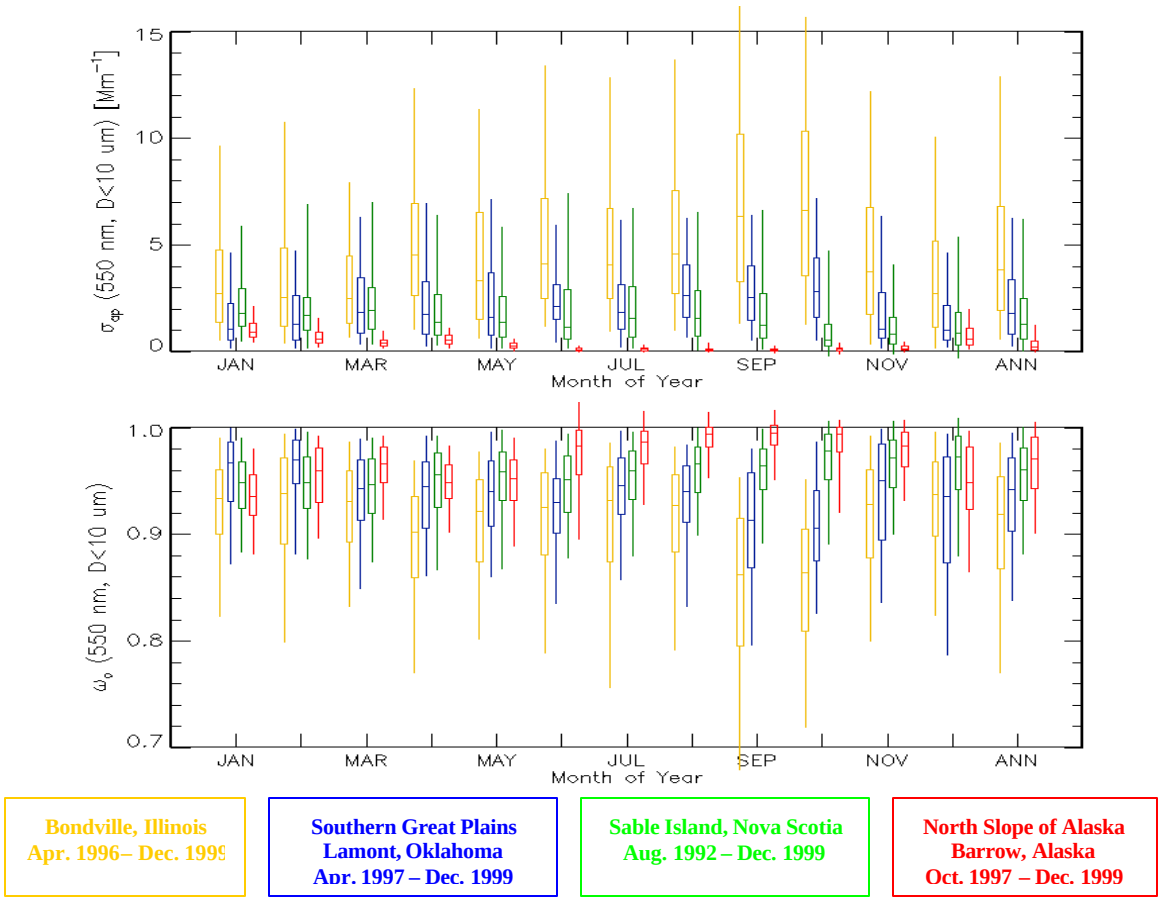

Figure 4. Monthly distributions of σ_{ap} and ω_0 at the four CMDL North American aerosol-monitoring sites.

transportation activities, or may be due to changing atmospheric flow patterns. A decrease in σ_{ap} (and increase in ω_0) during the last three months of the year is observed over the length of the ~7.5 year record at WSA. Aerosol light absorption at NSA begins to rise in December, presumably with the start of the winter/spring Arctic haze season. This higher-absorption period lasts into May, when the seasonal circulation (i.e., transport) patterns change and precipitation returns to the area.

An interesting relationship was discovered between ω_0 and σ_{sp} for the four CMDL aerosol-monitoring stations. At higher values of σ_{sp}, the ω_0 is also typically higher. This is illustrated in Figure 5. Hourly-average ω_0 data from each station over the entire period of record shown in Figure 4 have been binned according to the value of the scattering coefficient (10 Mm^{-1} bins). The North American sites show an increase in ω_0 with increasing σ_{sp}. The site marked "Kaashidhoo" represents data collected during February and March 1999 at the Kaashidhoo Climate Observatory in the Republic of Maldives in the Indian Ocean. This station samples polluted air from the Indian subcontinent and Southeast Asia at this time of year. The trend in the Kaashidhoo data is very different from that at the North American stations, and the relationship between ω_0 and σ_{sp} appears to have a negative slope. We speculate that regional differences in source emissions and removal mechanisms for absorbing versus nonabsorbing aerosols are responsible for this difference.

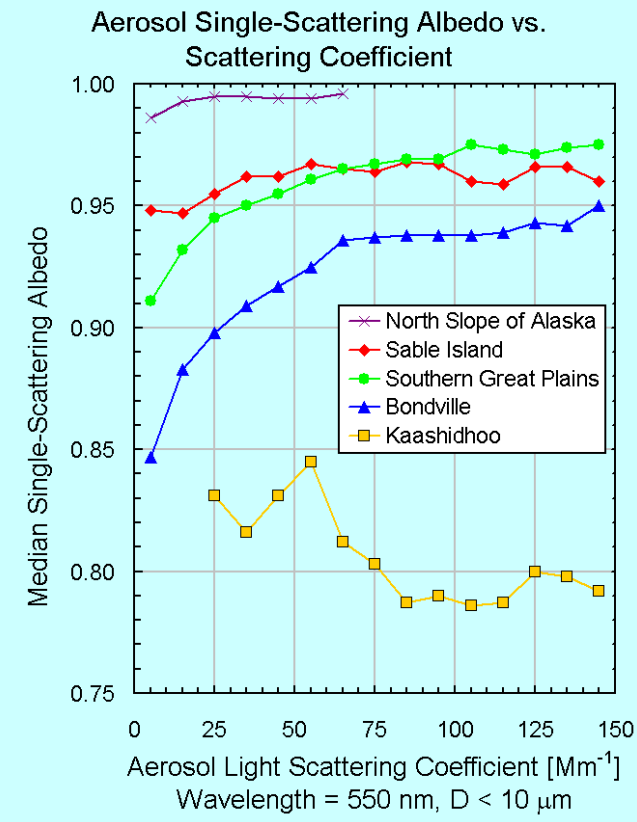

Figure 5. Relationship between ω_0 and σ_{sp} at four North American stations and one Indian Ocean surface aerosol-monitoring station.

Summary

The filter-based measurement of aerosol light absorption is a valuable tool for the determination of aerosol optical properties. The small size and relatively inexpensive cost of these absorption photometers make them the most widely used instruments for the determination of σ_{ap}. These absorption photometers are used throughout the NOAA CMDL aerosol-monitoring network, including the two located at ARM CART sites. Significant corrections are, however, required when using these filter-based techniques to estimate aerosol light absorption in the atmosphere.

Annual cycles in σ_{ap} and ω_0 are observed at the four CMDL North American aerosol monitoring sites, which include the SGP and NSA CART sites. The highest σ_{ap} and lowest ω_0 values for the SGP site occur during the months of September and October. This may be related to changes in regional scale agricultural or transportation activities, or to changes in atmospheric flow patterns. A similar cycle is seen at BND. NSA shows lower ω_0 values in the December to May period, which coincides with the Arctic haze season.

At the North American sites, periods showing the highest scattering coefficients also tend to show high single-scattering albedos. This suggests that the highest aerosol concentration episodes are driven by increases in scattering aerosols more so than increases in absorbing aerosols. The Indian Ocean station does not show this relationship, showing instead a slight tendency toward lower albedos associated with higher scattering episodes.

References

Anderson, T. L., and J. A. Ogren, 1998: Determining aerosol radiative properties using the TSI 3563 integrating nephelometer. *Aerosol Sci. Technol.*, **29**, 57-69.

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.