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Aerosol absorption measurements at Barrow, Mauna Loa and the south pole

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Abstract. Aerosol absorption (σ_{ap}) has been measured continuously using aethalometers at Barrow, Alaska (1986 to present); Mauna Loa, Hawaii (1990 to present); and south pole, Antarctica (1987-1990). These three stations are part of a network of baseline monitoring stations operated by the Climate Monitoring and Diagnostics Laboratory (CMDL) of the National Oceanic and Atmospheric Administration (NOAA). Condensation nucleus (CN) concentration and multiwavelength aerosol scattering (σ_{sp}) have also been measured continuously for many years at these stations. Aethalometer measurements are usually reported in terms of atmospheric black carbon aerosol (BC) concentration using the calibration suggested by the manufacturer. Here we deduce the in situ σ_{ap}(550 nm) from aethalometer measurements by assuming that the aerosol absorption on the aethalometer filter is enhanced by a factor of 1.9 over that in the atmosphere. This is consistent with using 19 $m^2 g^{-1}$ for the specific absorption of BC on the aethalometer filter and 10 $m^2 g^{-1}$ for the in situ specific absorption of BC in the atmosphere (the ratio of the two specific absorptions is 1.9). Although these values of specific absorption may vary significantly for different environments, the ratio might be expected to be relatively constant. The single-scattering albedo, defined by $\omega = \sigma_{sp}/(\sigma_{sp} + \sigma_{ap})$, has been estimated from the simultaneous measurements of σ_{ap} and σ_{sp}. Furthermore, assuming a $1/\lambda$ dependence for σ_{ap} in the 450 to 700-nm wavelength region, multiwavelength σ_{sp} measurements allow the estimation of the wavelength dependence of ω. Each station shows a considerable annual cycle in σ_{ap}, σ_{sp}, and ω. The maximum in the Barrow annual cycle is caused primarily by the springtime Arctic haze phenomenon; the maximum in the Mauna Loa annual cycle is caused by the springtime Asian dust transport; and the maximum in the south pole annual cycle is caused by late winter transport from southern midlatitudes. It was found that annual mean values are $\sigma_{ap} = 4.1 \times 10^{-7}$ m^{-1} (≈41 ng m^{-3} BC) and $\omega = 0.96$ for Barrow; $\sigma_{ap} = 5.8 \times 10^{-8}$ m^{-1} (≈5.8 ng m^{-3} BC) and $\omega = 0.97$ for Mauna Loa; and $\sigma_{ap} = 6.5 \times 10^{-9}$ m^{-1} (≈0.65 ng m^{-3} BC) and $\omega = 0.97$ for south pole. It was also found that the wavelength dependence of ω may be important at Barrow and south pole, but not important at Mauna Loa.

Introduction

Aerosol particles suspended in the atmosphere influence the Earth's climate in a direct or indirect manner. Direct effects refer to the interaction of the suspended particles with the radiation field in the atmosphere. Indirect effects refer to the effects of particles on cloud processes, which in turn affect climate. Possible climate forcing by anthropogenic aerosols has been discussed by *Charlson et al.* [1992] and *Kiehl and Briegleb* [1993]. Current climate forcing by anthropogenic sulfate is thought to be of comparable magnitude to that by greenhouse gases but is opposite in sign and is caused by aerosol backscattering to space. Because aerosol absorption may also be important, particularly on a local scale, it is important to establish current background values and to assess the possible climatic effects of anthropogenic contributions. *Clarke and Charlson* [1985] discussed the role of black carbon (BC) aerosol related to the background radiative properties of the atmosphere.

The Climate Monitoring and Diagnostics Laboratory (CMDL) (formerly GMCC) of the National Oceanic and Atmospheric Administration (NOAA) operates baseline atmospheric monitoring stations at Barrow, Alaska (BRW); Mauna Loa, Hawaii (MLO); American Samoa (SMO); and Amundsen-Scott Station, south pole (SPO). The mission of this program is to conduct measurements of atmospheric quantities that may influence the Earth's climate, to identify both natural and anthropogenic sources and sinks of these quantities, and to understand their possible effects on climate.

The CMDL baseline aerosol monitoring program consists of the continuous measurement of condensation nucleus (CN) concentration and aerosol scattering coefficient (σ_{sp}) at BRW, MLO, SMO, and SPO [*Bodhaine,* 1983]. The Importance of Carbon Particles in the Atmosphere Workshop, held in Boulder, Colorado, stressed the importance of absorbing particles and recommended that aerosol absorption coefficient (σ_{ap}) be measured at the CMDL observatories [*Bodhaine and Winchester,* 1981]. Continuous σ_{ap} measurements were initiated at BRW in 1986, MLO in 1990, and SPO in 1987 [*Bodhaine et al.,* 1989, 1992; *Hansen et al.,* 1988]. Short-term BC experiments at these stations were reported by *Rosen et al.* [1981], *Clarke and Charlson* [1985], *Bodhaine et al.* [1988], *Bodhaine* [1989a, b], and *Hansen et al.* [1989].

The purpose of this report is to summarize the σ_{ap} measurements made at the CMDL observatories and to present initial

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analyses for these stations. Simultaneous measurements of σ_{sp} and σ_{ap} allow the calculation of single-scattering albedo, $\omega = \sigma_{sp}/(\sigma_{sp} + \sigma_{ap})$, an important parameter in climate modeling.

Instrumentation

CN Counters

CN concentrations were originally measured at CMDL stations using General Electric (GE) automatic CN counters [*Bodhaine,* 1983]. The GE counter uses an expansion chamber to produce a cloud at a water supersaturation of about 300%. The density of the cloud is measured by optical means and converted to nuclei concentration through a calibration derived from daily comparisons with a Pollak CN counter [*Pollak and Metnieks,* 1960; *Bodhaine and Murphy,* 1980]. Thermo Systems Incorporated (TSI) CN counters (model 3760) were installed at BRW in March 1990, MLO in May 1988, and SPO in January 1989. The TSI counter is a continuous flow instrument that uses butyl alcohol as its working fluid to produce a cloud of alcohol droplets by directly cooling the airstream. The droplets are then counted by a single-particle counter to give the absolute concentration. The operation of the TSI counters was described by *Thermo Systems Incorporated (TSI)* [1987]. CN concentration data are used primarily to screen the σ_{sp} and σ_{ap} data for local pollution effects. A complete CN climatology is available for each station from the author.

Nephelometer

The σ_{sp} has been measured at the CMDL baseline stations using four-wavelength nephelometers with optics similar to that described by *Ahlquist and Charlson* [1969]. A continuous flow of air through the instrument is illuminated by a quartz-halogen lamp, and scattered light is detected by a photomultiplier (PMT). A rotating filter wheel in front of the PMT allows the detection of light at the nominal 450-, 550-, 700-, and 850-nm wavelengths. The instrument automatically switches between ambient air and filtered air and does a real-time subtraction to eliminate the instrument background (including the Rayleigh scattering of air). This instrument, described in detail by *Bodhaine* [1982], is capable of measuring σ_{sp} values as low as about 10^{-7} m^{-1}, about 1% of the scattering by air molecules.

Multiwavelength nephelometer data are often expressed as a power function of wavelength (λ) in the form $\sigma_{sp} = K_1\lambda^{-\alpha}$, where K_1 is a constant and α is known as the Ångström exponent. Aerosol size distribution information may be derived from multiwavelength nephelometer measurements [*Thielke et al.,* 1972] by examining α. In general, a large α implies smaller particles, and a small α implies larger particles.

Aethalometer

The light-absorbing component of the aerosol is measured at CMDL baseline stations with aethalometers manufactured by Magee Scientific [*Hansen et al.,* 1982]. The aethalometer measures the real-time attenuation of light transmitted through particles that accumulate on a quartz-fiber filter. A vacuum pump draws air through the instrument so that particles continuously accumulate on the filter while being illuminated by a lamp. Light transmission through the filter is sensed by two photocells: one through a sample spot where aerosol accumulates and another through a reference portion of the filter where no aerosol accumulates. The intensities of the two light beams are measured as a function of time, and the instantaneous attenuation of the sample beam may be recovered by taking the ratio of the sample beam to the reference beam and differencing the time series. The reference beam compensates for any variations in lamp brightness or drift in the electronics. Alternating periods of lamp-on and lamp-off allow for subtraction of detector dark currents. The instrument has an incandescent light source and a broadband detector; the spectral response for elemental carbon particles peaks at approximately 830 nm (A.D.A. Hansen, personal communication, 1991). Our instruments operate at a flow rate of about 11 standard liters per minute (slpm), and aerosol is accumulated on a circular spot with an area of about 0.95 cm^2.

The stability of the aethalometer optics allows detection of changes in transmitted light intensity of 1 part in 10^4, corresponding to a σ_{ap} noise level of about 1.5×10^{-8} m^{-1} (about 1.5 ng m^{-3} BC) for a 1-hour collection period. Instrument control and data acquisition are performed by dedicated laptop computers, and all data are available in real time and stored on floppy disks with a time resolution of 1 hour, with the exception of BRW where 10-min data are stored.

BC is usually the dominant absorber in the atmospheric aerosol [*Rosen et al.,* 1978]. Hematite (Fe_2O_3), a constituent of desert dust, is the only other strong absorber found commonly in the atmospheric aerosol; however, about 200 times as much hematite (mass) as BC is needed for equivalent absorption. *Schnell et al.* [1994] studied the relative contributions of dust and BC to σ_{ap} at MLO. They found a maximum of about 20% absorption contributed by dust during a large dust storm recorded at MLO. Similar results were found even in dust storms in the former Soviet Central Asia [*Hansen et al.,* 1993]. However, the contribution to absorption by dust at MLO is usually negligible.

Since the aethalometer by its very nature measures aerosol absorption on the filter, σ_{ap} may be inferred directly from light absorption on the filter by using a suitable calibration constant. In this work, σ_{ap} (per meter) was calculated using the formula

$$\sigma_{ap}(550\,\text{nm}) = -\frac{1}{1.9}\,\frac{A\ln(I_2/I_1)}{Q(t_2 - t_1)}, \qquad (1)$$

where I_1 and I_2 are the ratios of the intensities of the sample beam to the reference beam at times t_1 and t_2 (seconds); Q (cubic meters per second) is the volume flow rate of air through the filter; A (square meters) is the area of the spot on the filter; and the constant 1.9 is the enhancement of aerosol absorption for the aerosol embedded in the quartz-fiber filter matrix over that in the atmosphere, with the particular illumination used in the instrument. For simplicity the correction for detector dark current is not shown here in equation (1).

The aethalometer manufacturer provides a calibration factor that relates the attenuation of the light beam through the filter to the BC mass loading on the filter. To provide this calibration, BC loadings on filters collected in various locations were determined by measuring the CO_2 evolved in a temperature-programmed furnace. The manufacturer recommends that the BC concentration (grams per cubic meter) be calculated from the formula

$$\text{BC} = -\frac{1}{19}\,\frac{A\ln(I_2/I_1)}{Q(t_2 - t_1)}. \qquad (2)$$

However, equation (2) requires an assumption not needed in equation (1), that the value of the aethalometer specific absorp-

tion (19 $m^2 g^{-1}$) does not vary for different sources, size distributions, and mixtures of BC. *Liousse et al.* [1993] suggest a range of 5-20 $m^2 g^{-1}$ for the aethalometer specific absorption, which could introduce an error in estimates of BC. Here we wish to estimate σ_{ap} directly, and as noted above, the aethalometer actually measures aerosol absorption on the filter substrate (not BC).

Traditionally, estimating σ_{ap} from aethalometer measurements has been a two-step process. First, BC concentration is calculated using equation 2. Second, σ_{ap} is calculated using the equation $\sigma_{ap} = (S_a)(BC)$, where S_a is the specific absorption of BC suspended in the atmosphere. Commonly used values for S_a are of the order of 10 $m^2 g^{-1}$ at a wavelength of 550 nm [*Gerber,* 1982; *Clarke and Charlson,* 1985; *Bodhaine et al.,* 1992]. However, since the aethalometer measures σ_{ap} directly, the intermediate step of calculating BC is unnecessary. Rather, σ_{ap}(550 nm) can be calculated directly from aethalometer data using equation (1).

The constant 1.9 in equation (1) is consistent with the ratio of an aethalometer specific absorption of 19 $m^2 g^{-1}$ on the filter substrate and a specific absorption of 10 $m^2 g^{-1}$ at 550 nm for BC aerosol suspended in the atmosphere. Since the intermediate step of calculating BC has been eliminated, possible errors in the assumed value of 19 in equation (2) are avoided. It is likely that the value 1.9 in equation (1) remains fairly constant over a wide range of environmental conditions because in one sense it represents the ratio of the aethalometer instrumental specific absorption to the in situ atmospheric specific absorption. Although each might be expected to vary under different environmental conditions, they would be expected to vary similarly. Alternatively, the aethalometer enhances the apparent aerosol absorption by a factor of 1.9 (at 550 nm) because the particles are embedded in the filter substrate matrix. This factor of 1.9 relationship may also be inferred from the comparisons of *Rosen et al.* [1978], *Lin et al.* [1973], and *Sadler et al.,* [1981], as discussed by *Bodhaine et al.* [1992]. Further investigations are needed to verify that the ratio of the two specific absorptions is indeed fairly constant for different environments and that 1.9 is a suitable estimate for this ratio.

In this work, σ_{ap}(550 nm) was calculated from equation (1), and under the above assumptions, σ_{ap}(550 nm) = 1×10^{-8} m^{-1} is approximately equivalent to BC = 1 ng m^{-3}. This provides a convenient estimate of BC but does not take into account the contribution of absorbers other than BC (such as in a strong dust event at MLO).

Because σ_{ap} is approximately inversely proportional to λ for BC aerosol over the visible band [*van de Hulst,* 1957; *Jennings and Pinnick,* 1980; *Gerber,* 1982; *Horvath,* 1993; *Liousse et al.,* 1994; *Dobbins et al.,* 1994], it is possible to specify S_a as a function of wavelength and therefore to estimate σ_{ap} as a function of wavelength from the aethalometer data. Replacing the constant 1.9 by $C(\lambda)$ in the denominator of equation (1), and assuming that C(550 nm) = 1.9, we have for the purposes of this report that C(450 nm) = 1.5, C(550 nm) = 1.9, and C(700 nm) = 2.4. Note that σ_{ap} decreases with increasing wavelength. Finally, ω as a function of wavelength can be calculated from the corresponding multiwavelength σ_{ap} and σ_{sp} data. All σ_{ap} and σ_{sp} data are presented in volume units at the effective altitude of the particular observatory.

To examine the variation of ω with wavelength, consider that $\sigma_{sp} = K_1\lambda^{-\alpha}$ and $\sigma_{ap} = K_2\lambda^{-1}$, where K_1 and K_2 are constants. Then $\omega = K_1/(K_1 + K_2\lambda^{(\alpha-1)})$. Obviously, ω is independent of λ if $\alpha = 1$. However, ω will vary somewhat with λ for typical atmospheric values of α. For example, typical background values of α at BRW are about 1.5 [*Bodhaine,* 1989a], typical values at MLO are about 0-2 depending on the presence of Asian dust [*Bodhaine et al.,* 1992], and typical values at SPO are about 1.3-1.8 [*Bodhaine et al.,* 1986].

Data Analysis

Data Screening Techniques

To construct a data set consisting of screened background values of σ_{sp}, σ_{ap}, α, and ω, it was necessary to merge the σ_{sp}, σ_{ap}, wind direction (WD), and wind speed (WS) data sets to guarantee that background σ_{sp} and σ_{ap} were averaged over exactly the same time periods. Hourly average data sets were used as starting points for all subsequent averages. First, an hour of σ_{sp} data was accepted if data for 450, 550, and 700 nm were not missing. Only these three wavelengths of data were chosen because the data for 850 nm tend to be missing more often than the others. For Mauna Loa, because of a diurnal wind flow cycle causing exposure to local pollution sources, only nighttime data were accepted (nighttime data were defined as 2200-0800 Hawaii standard time (HST)). Next, if local pollution was suspected, that hour of data was excluded. Wind data for that hour were checked and that hour of data was excluded if WD was not from the station clean-air sector or if WS was < 0.5 m s^{-1}. The clean-air sectors used in this study for the three stations are given in Table 1. Finally, if the hourly data value for σ_{ap} was not missing, then σ_{sp}, σ_{ap}, α, and ω for that hour were included in 2-day mean calculations; α was calculated using the formula

$$\alpha = -\frac{1}{2}\left\{\frac{\log[\sigma_{sp}(450)/\sigma_{sp}(550)]}{\log(450/550)} + \frac{\log[\sigma_{sp}(550)/\sigma_{sp}(700)]}{\log(550/700)}\right\}, \quad (3)$$

where equation (3) is simply the average of the two independent values of α calculated for the intervals 450-550 and 550-700 nm, respectively; ω was calculated from the formula $\omega = \sigma_{sp}/(\sigma_{sp} + \sigma_{ap})$. Thus σ_{sp} and σ_{ap} were handled in identical manners for exactly the same time periods, ensuring the most accurate estimates of α and ω. Note that in cases when the various data sets had significant missing data, the merge process could result in small numbers of values available to calculate a given 2-day mean.

Two-day means were chosen to assemble a basic set of independent data points because hourly aerosol data at BRW are autocorrelated out to about 35-50 hours [*Bodhaine and Dutton,* 1993]. Therefore a 2-day mean is a convenient choice, and the same criteria were used for all three stations in this study. Once a set of 2-day mean data was constructed, monthly medians and

Table 1. Clean Air Sampling Sectors for BRW, MLO, and SPO

Station	Clean Air Sector
BRW	0° – 130°
MLO	90° – 270°
SPO	330° – 110°

BRW, Barrow, Alaska; MLO, Mauna Loa, Hawaii; SPO, south pole, Antarctica.

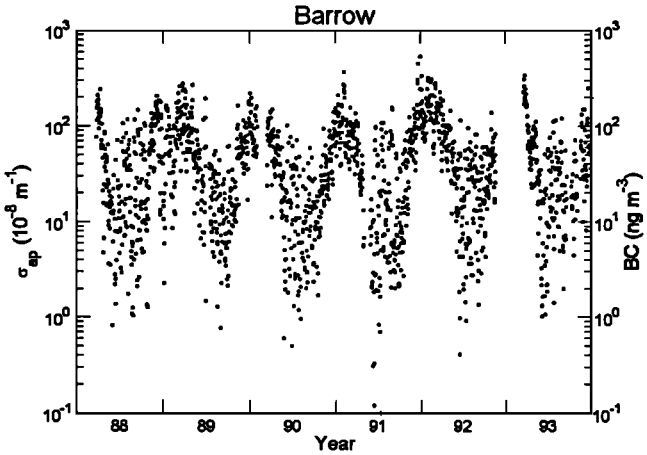

Figure 1. Daily mean black carbon (BC) aerosol concentration at Barrow, Alaska, for 1988-1993. No data screening or merging operations were performed.

quartiles for all data sets were calculated in order to study possible annual cycles at the three sites.

Barrow, Alaska

The Barrow monitoring station was established in 1972 at Point Barrow, the northernmost tip of Alaska. Aerosol measurements were initiated in 1976. This site has been found to be representative of the Arctic region and the springtime phenomenon known as the Arctic haze. The observatory building is located to the east of Barrow village and, with prevailing winds from the ENE, is upwind of local pollution sources about 76% of the time. A map of the area and a discussion of the influence of local pollution sources were presented by *Bodhaine et al.* [1981a]. CN and σ_{sp} data for Barrow were presented by *Bodhaine* [1983], *Murphy and Bodhaine* [1980b], and *Quakenbush and Bodhaine* [1986].

An aethalometer was first installed at BRW for ground-based σ_{ap} measurements during the second Arctic Gas and Aerosol Sampling Program (AGASP II) in March-April 1986. Results of this experiment were published in a special issue of the *Journal of Atmospheric Chemistry, vol. 9,* pp. 1-397, 1989 [see e.g., *Bodhaine et al.,* 1989].

Daily means of the entire BRW σ_{ap} data set are presented in Figure 1 in order to show the characteristics of the data set before merging and screening were performed. A repeatable annual cycle is evident with a maximum in March-April and a minimum in the late summer. The spring maximum is attributed to Arctic haze, an Arctic-wide phenomenon [*Joranger and Ottar,* 1984] caused by the long-range transport of anthropogenic pollution generated in the northern midlatitudes. Maximum values for σ_{ap} in the spring are about 3×10^{-6} m^{-1} (BC $\approx$ 300 ng m^{-3}) and minimum values are in the vicinity of 10^{-8} m^{-1} (BC $\approx$ 1 ng m^{-3}). Occasional clean values may be as low as $\sigma_{ap} = 2 \times 10^{-9}$ m^{-1} (BC $\approx$ 0.2 ng m^{-3}), comparable to clean conditions at MLO and SPO.

Monthly medians of the BRW σ_{sp}, σ_{ap}, and ω data at 550 nm are shown in Figure 2. The top and bottom bars show quartiles. Note that these monthly medians were constructed from hourly data according to the screening criteria discussed in the previous section, so that σ_{sp} and σ_{ap} were averaged over exactly the same time periods. In so doing, however, missing data in the various data sets can reduce the number of data points used to form the various medians and quartiles.

Referring to the σ_{sp} data in Figure 2, the annual cycle due to the Arctic haze phenomenon is apparent, with springtime monthly medians exceeding 2×10^{-5} m^{-1} and summertime monthly medians of about 10^{-6} m^{-1}. The σ_{ap} data show a similar annual cycle with a maximum of about 2×10^{-6} m^{-1} (BC $\approx$ 200 ng m^{-3}) and a minimum of about 7×10^{-8} m^{-1} (BC $\approx$ 7 ng m^{-3}). Because of the slightly stronger annual cycle in σ_{sp} a similar annual cycle is seen in ω with a minimum in springtime, suggesting that enhanced concentrations of BC coincide with Arctic haze events. Springtime ω monthly values are as low as about 0.9, indicating significant absorption, whereas summer values are as high as 0.98. Note that the July 1988 data point for ω has an upper quartile exceeding 1. The corresponding median for σ_{ap} is close to zero with an upper quartile of about 10^{-7} m^{-1}. These values of σ_{ap} could not be plotted on the graph but impossible values can occur in the calculations because of noisy data during clean conditions. However, the monthly median and upper quartile of σ_{ap} for July 1988 probably give reasonable limits on its actual value.

The monthly medians of the 1989 data shown in Figure 2 are presented in Table 2. To save space, the year with the best data recovery was chosen for each station. The peak of the Arctic haze event occurs in March, showing $\sigma_{sp} = 1.47 \times 10^{-5}$ m^{-1} and $\sigma_{ap} = 1.31 \times 10^{-6}$ (BC $\approx$ 131 ng m^{-3}). The corresponding α = 1.29 (maximum for the year) and ω = 0.928 (minimum for the year) suggest smaller and more absorbing particles, probably combustion aerosol, during that month. It should be noted that an ω value shown in the table is the median of the 2-day means for that month. This value can be slightly different than a value of ω calculated directly from the monthly medians of σ_{sp} and σ_{ap}.

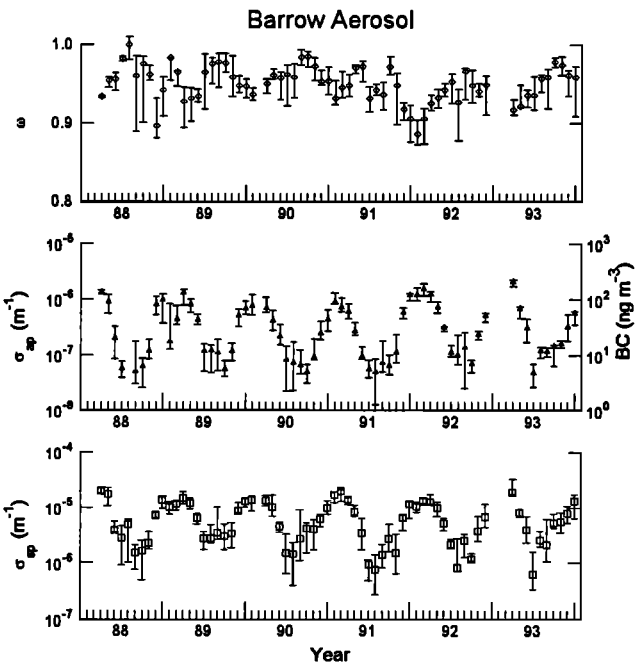

Figure 2. Monthly medians of σ_{sp}(550 nm), σ_{ap}, and ω at Barrow, Alaska, for 1988-1993. The upper and lower bars show the quartile points, respectively, for each month. Monthly medians were calculated from 2-day means according to the screening and merging criteria described in the text. The single-scattering albedo was calculated from $\omega = \sigma_{sp}/(\sigma_{sp} + \sigma_{ap})$ for each 2-day mean value.

Table 2. Monthly Medians of σ_{sp}(550 nm) (10^{-6} m^{-1}), α, σ_{ap}(550 nm) (10^{-8} m^{-1}), and ω for 1989 at BRW

Month	σ_{sp}	α	σ_{ap}	ω
Jan.	10.2	1.02	17.6	0.983
Feb.	11.6	1.04	43.6	0.965
March	14.7	1.29	131.0	0.928
April	11.8	1.26	79.6	0.932
May	6.48	1.62	42.8	0.934
June	2.74	1.84	11.7	0.965
July	2.76	0.67	12.0	0.976
Aug.	3.48	0.59	11.0	0.978
Sept.	3.04	0.16	5.6	0.977
Oct.	3.36	0.59	11.6	0.959
Nov.	8.51	0.95	51.2	0.948
Dec.	13.0	1.22	69.3	0.947
Mean	7.64	1.02	40.6	0.958

Mauna Loa, Hawaii

MLO is located at an altitude of 3.4 km on the north slope of Mauna Loa mountain, on the island of Hawaii. This site is often representative of the midtroposphere in that region of the Pacific Ocean, particularly during the nighttime downslope wind conditions [*Mendonca,* 1969]. Ordinarily, winds with a southerly component occur during the night and are representative of the background midtroposphere, and winds with a northerly component occur during the day but may be contaminated by local island processes. CN and σ_{sp} data for MLO were presented by *Bodhaine and Mendonca* [1974], *Bodhaine* [1978, 1981, 1983], and *Massey et al.* [1987].

The routine aerosol monitoring program at MLO was initiated in 1974 and consists of the measurement of CN concentration and σ_{sp}. Continuous σ_{ap} measurements were initiated at MLO in April 1990 using an aethalometer. *Bodhaine et al.* [1992] presented the first year of continuous σ_{ap} measurements at MLO. Measurements of σ_{ap} were made in 1982 by *Clarke and Charlson* [1985].

The well-known annual cycle in aerosol concentrations at MLO was discussed by *Bodhaine et al.* [1981b]; the springtime maximum is caused by the long-range transport of dust from Asian deserts [*Darzi and Winchester,* 1982; *Shaw,* 1980]. Trajectories calculated backward from MLO [*Miller,* 1981; *Harris and Kahl,* 1990] show frequent long-range transport from Asia during winter and spring. The aerosol maximum in April and May at MLO occurs when strong westerly flow coincides with dust storms over Asian deserts [*Merrill et al.,* 1989].

Daily means of the entire MLO σ_{ap} data set are presented in Figure 3. No merging or screening has been done, other than to include only nighttime data (2200-0800 HST). A repeatable annual cycle is evident with a maximum in April-May and a minimum in winter. As mentioned above, the springtime maximum is caused by the long-range transport of desert dust from Asia, suggesting that anthropogenic materials originating in the source regions are transported as well. Maximum values for σ_{ap} in the spring are about 10^{-6} m^{-1} (BC $\approx$ 100 ng m^{-3}) and minimum values are in the vicinity of 2×10^{-8} m^{-1} (BC $\approx$ 2 ng m^{-3}). Occasional clean values may be less than 10^{-9} m^{-1} (BC $\approx$ 0.1 ng m^{-3}), comparable to the cleanest conditions at SPO.

Monthly medians of the MLO σ_{sp}, σ_{ap}, and ω data at 550 nm are shown in Figure 4. The top and bottom bars show quartiles. Again, note that missing data in the various data sets can reduce the number of data points used to form the various medians and quartiles. Referring to Figure 4, the σ_{sp} data show an annual cycle with a maximum in April-May, caused by the long-range transport of Asian desert dust. The maximum monthly median of σ_{sp} is about 6×10^{-6} m^{-1} and the minimum is about 6×10^{-7} m^{-1}, giving an annual variation of about an order of magnitude. On the other hand, σ_{ap} varies from a maximum of about 3×10^{-7} m^{-1} (BC $\approx$ 30 ng m^{-3}) to a minimum of about 10^{-8} m^{-1} (BC $\approx$ 1 ng m^{-3}). This produces an annual cycle in ω that is in phase with those of σ_{sp} and σ_{ap} and varies between about 0.91 and 0.99. Evidently, BC of anthropogenic origin is transported from Asia along with the dust during the springtime season. Aerosol episodes at MLO were examined on a daily basis [*Bodhaine et al.,* 1992] and it was found that large values of σ_{sp} and BC usually coincide but can occur separately during strong transport from Asia. As shown by *Schnell et al.* [1994], absorption by dust can account for as much as 20% of the total absorption during the strongest dust episode. Therefore estimates of BC during these episodes could be significantly in error.

The monthly medians for 1992 were chosen as the best year for data recovery and are presented in Table 3. The peak of the Asian dust event of $\sigma_{sp} = 4.01 \times 10^{-6}$ m^{-1} occurred in April, the peak of $\sigma_{ap} = 2.83 \times 10^{-7}$ (BC $\approx$ 28.3 ng m^{-3}) occurred in March, and the minimum of $\omega = 0.943$ occurred in March, indicating that the annual cycles of σ_{sp} and σ_{ap} may have been slightly out of phase. Examination of Figure 4 shows that σ_{sp} and σ_{ap} were in phase for two of the years and slightly out of phase for two of the years; ω shows a maximum of about 0.99 in December and a minimum of about 0.94 in March.

South Pole, Antarctica

The Amundsen-Scott Station was established at the geographic south pole for the International Geophysical Year in 1956; CMDL began measurements in 1974. The CMDL program is located upwind from the camp in the Clean Air Facility, constructed by the National Science Foundation on the greater polar plateau at about 2.84 km (mean sea level (msl)). The prevailing winds are from about grid NE and are from the clean-air sector about 95% of the time. *Bodhaine et al.* [1986] presented a map of the site and a general description of sampling conditions.

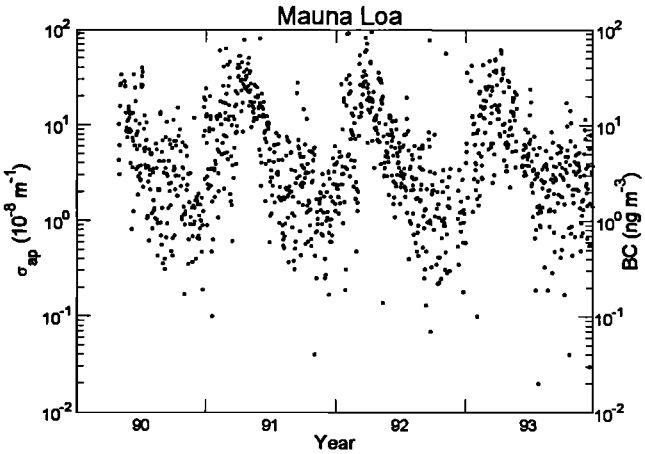

Figure 3. Daily mean BC aerosol concentration at Mauna Loa, Hawaii, for 1990-1993. No data screening or merging operations were performed.

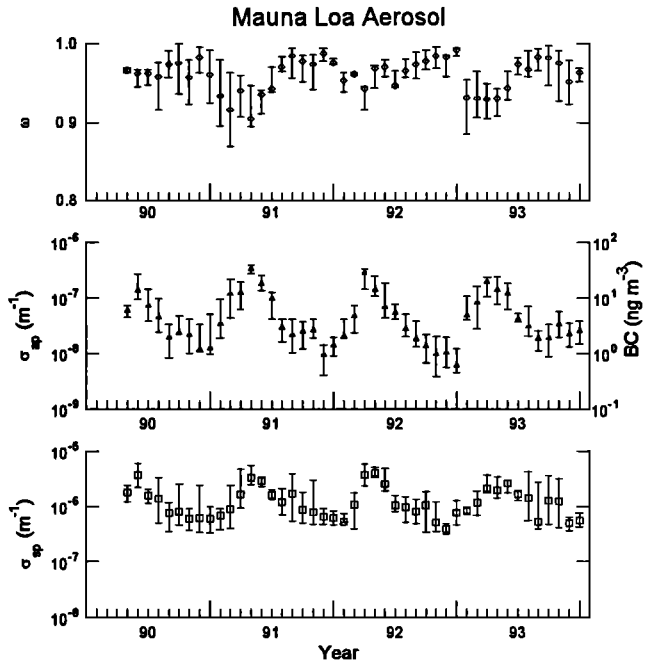

Figure 4. Monthly medians of σ_{sp}(550 nm), σ_{ap}, and ω at Mauna Loa Hawaii, for 1990-1993. The upper and lower bars show the quartile points, respectively, for each month. Monthly medians were calculated from 2-day means according to the screening and merging criteria described in the text. The single-scattering albedo was calculated from $\omega = \sigma_{sp}/(\sigma_{sp} + \sigma_{ap})$ for each 2-day mean value.

The routine aerosol measurement program began in 1974 with the installation of a Pollak CN counter and a GE automatic CN counter. A four-wavelength nephelometer was installed at SPO in 1979. CN and σ_{sp} data for SPO were presented by *Bodhaine and Murphy* [1980], *Murphy and Bodhaine* [1980a], *Bodhaine and Bortniak* [1981], *Bodhaine et al.* [1986, 1987], *Bodhaine and Shanahan* [1990], and *Bodhaine and Harris* [1992]. The first σ_{ap} measurements were made during 1987 and presented by *Hansen et al.* [1988], and measurements were continued through December 1990.

Table 3. Monthly Medians of σ_{sp}(550 nm) (10^{-6} m^{-1}), α, σ_{ap}(550 nm) (10^{-8} m^{-1}), and ω for 1992 at MLO

Month	σ_{sp}	α	σ_{ap}	ω
Jan.	0.525	1.77	1.99	0.954
Feb.	1.06	2.24	4.78	0.962
March	3.69	1.15	28.30	0.943
April	4.01	1.29	14.00	0.969
May	2.52	0.58	6.92	0.971
June	1.05	0.96	5.37	0.947
July	0.947	1.26	2.80	0.966
Aug.	0.790	1.60	1.78	0.974
Sept.	1.03	1.81	1.35	0.978
Oct.	0.507	1.64	0.97	0.985
Nov.	0.388	1.46	1.02	0.983
Dec.	0.762	1.50	0.59	0.992
Mean	1.44	1.44	5.82	0.969

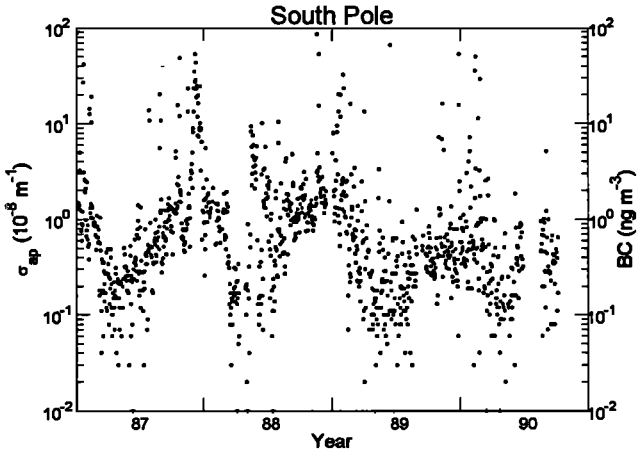

Figure 5. Daily mean BC aerosol concentration at Amundsen-Scott Station, south pole, for 1987-1990. No data screening or merging operations were performed.

Daily means of the entire SPO σ_{ap} data set are presented in Figure 5. Again, no merging or screening has been done. A repeatable annual cycle is evident with a maximum in the austral summer and a minimum in early winter. Maximum values for σ_{ap} in the summer are about 5×10^{-7} m^{-1} (BC $\approx$ 50 ng m^{-3}) and minimum values are in the vicinity of 2×10^{-10} m^{-1} (BC $\approx$ 0.02 ng m^{-3}). Occasional clean values may be of the order of 10^{-10} m^{-1} (BC $\approx$ 0.01 ng m^{-3}).

Monthly medians of the 1987 SPO σ_{sp}, σ_{ap}, and ω data at 550 nm are shown in Figure 6. The upper and lower error bars show quartiles. Because of missing data in the various data sets the

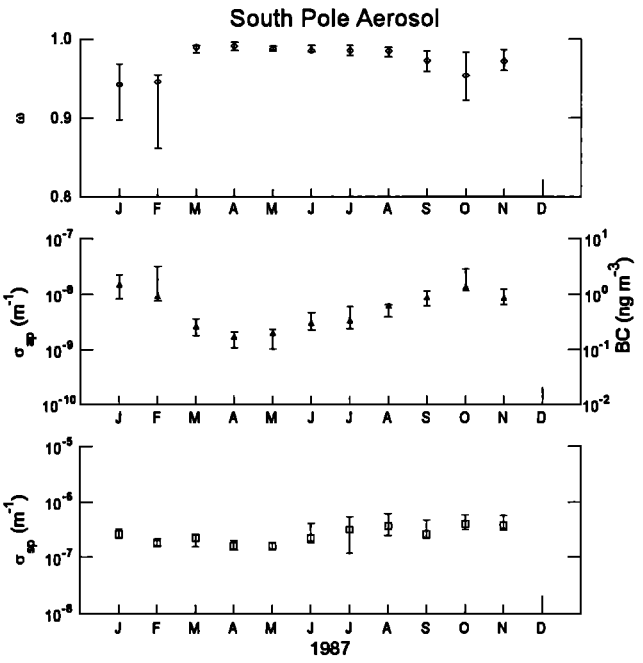

Figure 6. Monthly medians of σ_{sp}(550 nm), σ_{ap}, and ω at Amundsen-Scott Station, South Pole, for 1987. The upper and lower bars show the quartile points, respectively, for each month. Monthly medians were calculated from 2-day means according to the screening and merging criteria described in the text. The single-scattering albedo was calculated from $\omega = \sigma_{sp}/(\sigma_{sp} + \sigma_{ap})$ for each 2-day mean value.

Table 4. Monthly Medians of σ_{sp}(550 nm) (10^{-6} m^{-1}), α, σ_{ap}(550 nm) (10^{-8} m^{-1}), and ω for 1987 at SPO

Month	σ_{sp}	α	σ_{ap}	ω
Jan.	0.255	2.89	1.46	0.942
Feb.	0.180	3.19	0.90	0.946
March	0.222	2.38	0.25	0.990
April	0.161	2.27	0.17	0.991
May	0.158	2.33	0.20	0.988
June	0.219	1.96	0.30	0.986
July	0.314	1.75	0.32	0.986
Aug.	0.362	1.44	0.60	0.985
Sept.	0.256	1.54	0.86	0.973
Oct.	0.393	1.56	1.29	0.954
Nov.	0.376	2.05	0.82	0.972
Mean	0.263	2.12	0.65	0.974

merging procedure resulted in a limited number of data points in 1988-1990. Therefore monthly medians (and quartiles) are shown only for 1987. Referring to Figure 6, the σ_{sp} data show an annual cycle with a minimum in the late austral summer of about 10^{-7} m^{-1} and a maximum of about 4×10^{-7} m^{-1} in the late winter. The maximum has been identified as the long-range midtropospheric transport of sea salt from oceanic areas surrounding the Antarctic continent [*Bodhaine et al.*, 1986, 1987; *Parungo et al.*, 1981; *Harris*, 1992]. The ω data in Figure 6 show an annual cycle with a maximum in the winter and a minimum in the summer, suggesting that the sea-salt transport events do not contribute to enhanced concentrations of BC relative to σ_{sp}.

Table 4 presents the monthly medians for the data shown in Figure 6. As discussed above, the σ_{sp} and σ_{ap} data have somewhat similar annual cycles, but the resulting ω values have a significant annual cycle with a maximum in the austral winter and a minimum in the summer. Cross correlations were calculated for the data in Table 4. The strongest correlation is between BC and ω (−0.92), suggesting that BC controls ω, because the annual cycle of BC is stronger than that of σ_{sp}. The only other significant correlation was between σ_{sp} and α (−0.62). The smallest correlations were between α and BC (0.14) and σ_{sp} and ω (−0.19). The smallest value of α occurred as expected during August, the month of strongest sea-salt transport, suggesting larger particles.

Discussion

One of the goals of this paper is to estimate σ_{ap} and ω as a function of λ at the three stations, using multiwavelength nephelometer data (assuming that σ_{ap} varies as $1/\lambda$). For each station, two months were chosen (from Tables 2, 3, and 4), representative of extremes in the annual cycle of ω for that station. March and September 1989 were chosen for Barrow; April and December 1992 were chosen for Mauna Loa; and February and August 1987 were chosen for south pole. The results of these calculations are shown in Table 5. The σ_{sp}(450), σ_{sp}(550), σ_{sp}(700), and σ_{ap}(550) values shown in Table 5 are the monthly medians measured for each station/month at the given wavelength. The σ_{ap}(450) and σ_{ap}(700) values were calculated from the σ_{ap}(550) values assuming that σ_{ap} varies as $1/\lambda$; α_{12} and α_{23} were calculated from σ_{sp}(450) and σ_{sp}(550) and from σ_{sp}(550) and σ_{sp}(700), respectively. Finally, ω(450), ω(550), and ω(700) were calculated from the respective values of σ_{sp} and σ_{ap}.

It is apparent from Table 5 that σ_{sp} and σ_{ap} vary significantly with wavelength. (Please recall that $\sigma_{sp}(\lambda)$ was actually measured at 450, 550, and 700 nm, whereas $\sigma_{ap}(\lambda)$ was measured at 550 nm and assumed to vary as $1/\lambda$.) It is interesting to note that the monthly variation of σ_{sp}(550) and σ_{ap}(550) at the quartile points is about the same as the variation of σ_{sp} and σ_{ap} between adjacent wavelengths for all six cases shown in Table 5. This suggests that for monthly estimates of σ_{sp} and σ_{ap}, over the wavelength band 450-700 nm, variability with wavelength is not an important consideration at these three sites. The behavior of $\omega(\lambda)$ is interesting in light of the previous discussion (at the end of the aethalometer section); ω will vary with λ only if α differs significantly from 1, and ω can either increase or decrease with λ depending on whether $\alpha < 1$ or $\alpha > 1$. For example, ω for BRW in March increases and then decreases with λ, because $\alpha_{12} < 1$ and $\alpha_{23} > 1$. Similarly, ω for BRW in September simply increases with λ; ω for MLO does not vary significantly with λ because α is close to 1. On the other hand, ω for SPO in February decreases significantly with wavelength (from 0.96 to 0.90) be-

Table 5. Values of σ_{sp}, σ_{ap}, ω, and α at the 450-, 550-, and 700-nm Wavelengths for BRW During March and September 1989, MLO During April and December 1992, and SPO During February and August 1987

Station Year	Month	(10^{-6} m^{-1}) σ_{sp}450	σ_{sp}550	σ_{sp}700	(10^{-8} m^{-1}) σ_{ap}450	σ_{ap}550	σ_{ap}700	ω450	ω550	ω700	α_{12}	α_{23}
BRW	March	16.5	14.7	9.18	160.	131.	103.	0.922	0.928	0.911	0.58	1.95
1989	Sept.	2.97	3.04	2.65	6.82	5.58	4.38	0.971	0.977	0.979	−0.12	0.57
MLO	April	4.90	4.01	2.94	17.1	14.0	11.0	0.969	0.969	0.967	1.00	1.29
1992	Dec.	0.962	0.762	0.612	0.726	0.594	0.467	0.992	0.992	0.992	1.16	0.91
SPO	Feb.	0.304	0.180	0.0725	1.10	0.901	0.708	0.960	0.946	0.900	2.61	3.77
1987	Aug.	0.461	0.362	0.250	0.736	0.602	0.473	0.986	0.985	0.983	1.20	1.53

The value of α_{12} was calculated from σ_{sp}(450) and σ_{sp}(550); α_{23} was calculated from σ_{sp}(550) and σ_{sp}(700). Note that the units of σ_{ap} are expressed such that the values shown in the table for σ_{ap}(550) may be interpreted approximately as BC (nanograms per cubic meter).

cause $\alpha > 1$. In spite of these considerations, however, the monthly variation of $\omega(550)$ at the quartile points (for all three stations) is larger than the variation of ω between adjacent wavelengths, suggesting that the variation of ω with wavelength is not an important consideration (over 450-700 nm) for monthly estimates. This is because the monthly variabilities of σ_{sp} and σ_{ap} tend to be relatively large; however, wavelength may be important when considering variability on other timescales.

It is difficult to predict the climatic effects of the aerosols discussed in this report without a comprehensive model. Indeed, one of the purposes of this report is to provide aerosol parameters for incorporation into climate models. However, a few general comments can be made. As discussed by *Ackerman* [1988], two important parameters in aerosol climate models are ω and the albedo of the underlying surface. An $\omega \approx 0.85$ appears to be a critical value, such that $\omega < 0.85$ would warm the system and $\omega > 0.85$ would cool the system. Furthermore, a high surface albedo (bright surface, such as snow) will enhance warming, and a low albedo (dark surface, such as ocean) will enhance cooling. In all cases, weakly absorbing aerosol (large ω) will produce a cooling. Strongly absorbing aerosol will produce warming for a bright surface and cooling for a dark surface, depending on the values used in the model for ω and surface albedo. The surface albedo is high for BRW and SPO (ice and snow, about 0.85) and low for MLO (ocean, about 0.06). During summer in the Arctic, breakup of the icepack near BRW results in a lower surface albedo that could reduce a warming effect.

All of the monthly median values of ω reported here for all three sites are above 0.9, well up in the range that would be expected to produce cooling, regardless of the surface albedo. However, short-term measurements near Barrow have suggested heating rates during Arctic haze events as high as 0.16-0.25 K d^{-1} [*Valero and Ackerman*, 1986]. Thus significant warming can occur in the Arctic on a regional basis and over timescales of several days when lower values of ω occur. Long-term average values of ω for MLO reported by *Clarke and Charlson* [1985] and *Bodhaine et al.* [1992] are also in the range that would be expected to produce cooling. However, *Bodhaine et al.* [1992] showed daily average events of $\omega = 0.70$ and 0.85 during February 1991. The author has personally observed Asian dust events at MLO that nearly obscure the Sun. Evidently, warming events may be occasionally possible at MLO but have not been documented. No estimates of ω have previously been made for SPO. It is clear from the data presented here that the aerosol climatic effect at SPO would be a cooling but would be small.

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