MONITORING AEROSOL OPTICAL DEPTH AT BARROW, ALASKA AND SOUTH POLE; HISTORICAL OVERVIEW, RECENT RESULTS, AND FUTURE GOALS

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ABSTRACT

Atmospheric aerosols affect the Earth's radiation budget through interactions with solar and terrestrial radiation. Various committees involved with assessing global climate change recognize that aerosols can significantly impact the earth's radiation balance. In particular, the Scientific Committee on Antarctic Research has recommended the establishment of an international network of solar spectrophotometers to monitor aerosol optical depth (AOD) at high latitudes. Although such a network now exists, better coordination is needed in order to provide research quality data to the scientific community. The U.S. National Oceanic and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory (CMDL) is collaborating with other institutes to assimilate AOD data from all polar observatories into a central archive for analysis. Historically, in situ aerosol data have been collected at CMDL baseline observatories located near Barrow, Alaska (BRW) and at South Pole, Antarctica (SPO), and since January 2000 continuous photometric measurements (during sunlit periods) have been made at these sites. An overview of past and current CMDL efforts to monitor AOD is given and some recent results are presented. Significant differences between the magnitudes and spectral signatures of AOD measured at BRW and SPO highlight the importance of assimilating similar data sets from other locations to better characterize polar aerosols spatially and temporally. Initial efforts should focus on defining natural cycles of AOD at a number of high latitude sites. Once these cycles are understood, more accurate assessments of climate forcing due to anthropogenic aerosol perturbations are possible. Through international cooperation this work can be expedited.

1. INTRODUCTION

Atmospheric aerosols affect the Earth's radiation budget directly by scattering and/or absorbing solar and infrared radiation, and indirectly as cloud condensation and ice nuclei. While the impacts of aerosols are often pronounced downwind of mid-latitude industrial complexes (e.g., Charlson et al., 1992), high latitude regions are also sensitive to variations in aerosol concentrations. This is especially true in the Arctic when airborne pollutants, dust and smoke from Eurasia are transported poleward. And, whereas aerosols tend to have a cooling effect at low and mid latitudes, their radiative effects are less certain for polar regions. The effects of high surface albedo in conjunction with low solar incident angles may cause atmospheric warming instead (Cacciari et al., 2000). Further complications arise when considering variations in the chemical and physical makeup of aerosols and their inhomogeneous distributions.

In 1998, the Scientific Committee on Antarctic Research (SCAR) recognized the importance of characterizing polar aerosols and recommended the establishment of an international network of solar spectrophotometers to monitor aerosol optical depth (AOD) at high southern latitudes. Such a network now exists and some participating countries also maintain sites in the Arctic, thus providing a bi-polar perspective. The concerns of SCAR are echoed in a recent Intergovernmental Panel on Climate Change assessment (IPCC, 2001). The IPCC executive summary calls for an expansion of "the observational foundation for climate studies" and a better understanding of "factors leading to changes in radiative forcing; in particular, (to) improve the observations of the spatial distribution of greenhouse gases *and aerosols*."

NOAA established observatories for monitoring climate at South Pole (SPO; 90°S) and at Barrow, Alaska (BRW; 71.32°N, 156.61°W) in the mid 1970s. Many time series are now of sufficient length to detect trends, and are appropriate for studying the physical mechanisms that underlie climate change. Beginning in 1973 annual reports have been published by CMDL (formerly Geophysical Monitoring for Climate Change, GMCC) that summarize its research. Data can be accessed and descriptions of ongoing activities obtained at: <u>http://www.cmdl.noaa.gov</u>. The focus of the CMDL aerosol and radiation program is to characterize the means, variability, and trends of aerosol properties on a regional basis using data from its baseline observatories. A further goal is to understand the factors that control the distribution of aerosols and their radiative properties. The CMDL radiation group has established new projects at BRW and SPO to measure spectral AOD on a more continuous basis. These efforts will extend historical records and provide greater opportunities to evaluate the radiative effects of aerosols in polar regions.

2. HISTORICAL STUDIES OF AEROSOLS AT NOAA/CMDL POLAR OBSERVATORIES

2.1 Aerosol Optical Depth Observations at SPO and BRW

Optical depth, which is a measure of the total extinction of radiation that enters and passes through the atmosphere, is of critical importance to the understanding of how climate is forced radiatively. Since 1977 CMDL has monitored an *effective* visible (500 nm) aerosol optical depth (AOD_{eff}) at BRW and SPO. AOD_{eff} is derived from measurements made using pyrheliometers fitted with moderately wideband filters. The data are accurate to within ±0.04 optical depth units (Dutton and Christy, 1992). While the data are not collected continuously, the unusually long time series have been valuable for defining seasonal background values and for detecting perturbations caused by major volcanic events. **Figure 1** highlights the increases in AOD_{eff} at BRW and SPO that resulted from the dispersion of volcanic aerosols after the eruptions of El Chichon and Pinatubo in 1982 and 1991, respectively. After major eruptions volcanic aerosols are dispersed globally by

stratospheric winds and then decay exponentially. Using a limited number of spectral observations of aerosol extinction a preliminary understanding of the evolution of the size distribution of volcanic aerosols has been achieved (Stone et al., 1994; Stone et al., 1993). Decay times were found to be longer at BRW and SPO than at lower latitudes. The persistence of volcanic aerosols at high latitudes is attributed to processes that occur within the polar vortices. Aerosols persist if dynamical processes favor vaporization at high altitudes and regeneration of particles at lower altitudes (Hofmann and Rosen, 1987). Also, natural elimination processes may be suppressed because vertical mixing within vortices is minimal. Quantifying the radiative forcing by volcanic aerosols at high latitudes has not been possible, however, because continuous records needed for such analyses are not yet available.



Figure 1. Time series of effective visible aerosol optical depth (AOD_{eff}) derived from filtered pyrheliometer measurements made at Barrow, Alaska and at South Pole. Two-year smoothed values are shown to highlight the periods of increase and subsequent decay following the eruptions of El Chichon (1982) and Pinatubo (1991). The dashed lines represent the long-term average background conditions (data provided by E. Dutton of NOAA/CMDL).

2.2 In Situ Observations of Aerosol Properties at SPO and BRW

Historically, optical properties of aerosols have been measured at the surface at SPO and BRW since the mid 1970s using multi-channel nephelometers (Bodhaine, 1996; Bodhaine, 1992). From these in situ measurements, seasonal cycles of aerosols at these sites have been well established. Recent efforts have focused on correlating the chemical and physical properties of aerosols with atmospheric cycles of specific size-selected chemical species (Quinn et al., 2002; Ogren, 1995). Depending on season, the presence of Arctic haze, Asian dust, sea salt aerosols, and occasionally smoke from Siberian forest fires are apparent in the BRW aerosol record. Sea salt aerosols influence the SPO record whenever marine air is transported to the interior plateau by large storm systems. There is need for improved optical parameterizations of these diverse aerosol species for quantifying their radiative effects on climate.

Arctic haze, resulting from emissions of industrial pollution throughout Europe and Russia has long been a subject of considerable interest and concern (e.g., Barrie, 1986; Shaw and Khalil, 1989). In Alaska this phenomenon is observed every winter/spring as a consequence of long-range transport (e.g., Harris and Kahl, 1994). Bodhaine and Dutton (1993) reported a decreasing trend in Arctic haze at BRW that they attributed to diminished emissions of pollutants in Eurasia after the demise of the Soviet Union. However, a definitive explanation for the trend has not been given. While emission rates are certain to influence the distribution of haze, changes in atmospheric transport may also be a factor as suggested by Jaffe et al. (1995). In fact, atmospheric circulation in this region has undergone significant shifts in recent decades that have resulted in increased cloudiness and warmer temperatures, but diminished snowfall (Stone, 1997; Stone et al., 2002). In the future, closer attention should be given the role of transport as it affects the evolution of specific aerosol species; e.g., sea salt, haze, volcanic, etc., and also their interactions with different types of cloud systems.

At SPO, intrusions of sea salt aerosols are observed when storms reach the interior from surrounding oceanic regions (e.g., Bodhaine, 1992). Clear signatures distinguish air masses carrying coarse (sodium) and small (sulfate) particles, with peak sodium concentrations associated with flow from the Weddell and Ross sea regions. Bergin et al. (1998) related in situ aerosol measurements from South Pole with collated records of chemical species captured in firn samples. The possibility now exists to resolve, on a seasonal basis, a record of aerosol properties dating from 1000 years ago by identifying chemical signatures embedded in Antarctic ice cores. Moreover, the natural (e.g., seasalt, biogenic, and volcanic) signatures should be distinguishable from anthropogenic ones. These results are corroborated by the studies of Stortini et al. (2000), and Traversi et al. (2000) that explain the variability of Antarctic aerosols in terms of the transport of marine air into the interior of the continent. Although the direct radiative impact of such aerosols can be studied theoretically (e.g., Cacciari, et al., 2000), verification of results will require continuous monitoring of aerosol optical properties in conjunction with measurements of the surface radiation budget. In particular, properties derived from in situ measurements need to be compared with those derived from photometric techniques that integrate the radiative effects of aerosols through the entire atmospheric column. Multi-spectral AOD measurements can provide a basis for such verification.

3. RECENT ENHANCEMENTS OF SPO AND BRW RADIATION PROGRAMS

3.1 Sunphotometry at SPO and BRW

It is clear that a better characterization of polar aerosols is needed to address the concerns of the climate and remote sensing communities. CMDL has enhanced its monitoring programs at BRW and SPO to include continuous measurements of spectral AOD, during sunlit periods, using photometers fitted with narrowband interference filters. These programs were initiated in January 2000, and in March 2000 at SPO and BRW, respectively. Because background values of Arctic and Antarctic optical depths are very small (e.g., Figure 1), the determination of spectral AOD at polar sites requires extra care to produce high quality data sets. The main features of these polar supphotometer projects include:

• Deployment of 4-channel, sun-tracking photometers to measure extinction at nominal wavelengths centered at 412, 500, 675, and 862 nm (368, 412, 500 and 865 nm prior to July 2000)

- Acquisition of raw voltages at one-minute resolution
- Use of ancillary radiometric data to evaluate data quality
- Photometric data are merged with station pressure and temperature to account for Rayleigh scattering using the method of Bodhaine et al. (1999), and to correct airmass calculations for refraction at low sun angles
- Thermal control of instruments within $\pm 1^{\circ}$ C of established set point (20°C) to increase filter longevity and assure consistency of field and calibration measurements
- Pre- and post-season calibrations at Mauna Loa Observatory (MLO) using the Langley method (e.g., Shaw, 1983) are made to track filter performance and assure accurate derivation of AODs (see *Section 3.2*)
- Use of daily O₃ and NO₂ data to account for absorption by these gases (see *Section 3.3*)
- Data are carefully screened using an interactive, graphical editor to eliminate cloud contamination, and suspect data due to operational problems, tracking errors, etc.
- When processing the aureole sunphotometer (SP01-A) data, corrections are made to account for first order scattering due to diffuse radiation within the instrument's field of view

Figure 2 shows a typical deployment of a sun-tracking spectral radiometer used for deriving aerosol optical depth at BRW and SPO.



Figure 2. NOAA/CMDL 4-cahnnel aureole sunphotometer SP01-A mounted on a solar tracker at the Barrow Observatory. Behind is the suite of radiometers used to measure components of downwelling solar irradiance used in determining the net surface radiation budget.

3.2 The Key to Successful Sunphotometry: Frequent Calibrations

Of critical importance to any sunphotometer program is the routine calibration of each instrument under stable atmospheric conditions. CMDL is fortunate in that MLO is a superb calibration site, situated at a high elevation (3397 m), located remotely on the island of Hawaii (19.54°N, 155.58°W). Throughout the year, there are frequent mornings in which the atmosphere is very stable, clean, and cloud-free; ideal conditions for making observations over the range of solar zenith angles necessary to produce high quality Langley plots. The Langley method of calibration has been used historically by many investigators; e.g., Shaw (1983), Reddy et al. (1990), Tomasi et al. (1992), and CMDL continues this practice. **Figure 3** summarizes the method and illustrates the repeatability that is possible when a set of Langley plots is obtained at various sites.

SP02-1022 500nm calibrations 2001



Figure 3. Langley plots (at 500 nm) obtained at three locations, altitudes, and times of year for the NOAA/CMDL photometer SP02-1022. Note the superior repeatability of the Mauna Loa intercepts in comparison with Boulder's. The dashed curve is an example of a plot obtained at Boulder in which atmospheric turbidity varied during the observation period. All regressions shown were produced from data collected during morning hours, over an airmass range, 4.75 to 2.25.

The fact that the zero-airmass intercept is nearly invariant over time, at different sites, and under different aerosol loadings (as evidenced by the varying slopes), indicates the high precision of the photometers currently in use by CMDL. However, it is essential that conditions during any given observation sequence be stable which is most often the case at MLO, sometimes at BRW, but seldom in Boulder, Colorado. Even at South Pole it is possible to produce a Langley plot if data from several weeks is used but it is not a valid calibration because aerosol concentrations vary continuously. Although Langley plots are produced operationally at all sites to monitor instrument stability, only the MLO calibrations are used during data processing. Note that the mean voltage of extrapolated zero-airmass intercepts is the calibration constant used when evaluating solar attenuation at any specified wavelength λ . These constants represent the extraterrestrial values of solar flux $V_o(\lambda)$ (in units of volts) from which measured voltages $V(\lambda)$ are referenced to derive values of optical depth. To date, MLO calibration constants have varied by less than 0.2% of mean values at one standard deviation. Thus, very accurate optical depth retrievals are possible operationally.

3.3 Determinations of Spectral Aerosol Optical Depth

Optical depth, τ_{λ} , at a central wavelength λ , can be derived by inverting the following expression of the Bouguer-Lambert-Beer law (e.g., Vitale and Tomasi, 1990):

$$V(\lambda) = RV_{O}(\lambda)e^{-m\tau}\lambda$$
⁽¹⁾

where, $V(\lambda)$ is the measured voltage, R is a correction factor that accounts for variations in the sun-earth distance, m is the refracted path length through the atmosphere (or airmass) at the time when the measurement is made, and as noted above, $V_o(\lambda)$ is the calibration constant, or extraterrestrial flux in units of volts.

To obtain aerosol optical depth (AOD) from (1), it is necessary to account for attenuation by other constituents of the atmosphere, depending on wavelength. Rayleigh

scattering occurs at all wavelengths and is accounted for using the method of Bodhaine et al. (1999), while ozone, and nitrogen dioxide affect only certain channels. O_3 absorbs at 500 nm and 675 nm, and NO₂ at 412 nm and 500 nm. Component optical depths for O₃ and NO₂ are computed on the basis of daily average total column amounts of these gases scaled by their respective absorption coefficients. Ozone is measured a few times a day at SPO and BRW using Dobson spectrophotometers (Schnell et al., 2001). However, NO₂ is not measured at either site and must be estimated using climatological values derived from satellite data in conjunction with ground-based measurements made at nearby sites. To date, NO₂ derived from the Polar Ozone and Aerosol Measurement III (POAM III) instrument of the French Space Agency and the Global Ozone Monitoring Experiment (GOME) on board the second European Remote Sensing Satellite (ERS-2) have been used in conjunction with data from Arrival Heights, Antarctica to produce seasonal climatologies for SPO and BRW (Stone, unpublished results). The uncertainties in AOD due to errors in NO₂ concentrations have not been fully evaluated but are probably less than 10% of typical AOD values, assuming that seasonal variations of the gas concentrations are taken into account. Errors resulting from changing ozone concentrations are probably less than 10% because ozone is measured 2-3 times each day at the stations.

Thus, while the CMDL photometers have performed very well and are fairly precise, evaluations of AOD remain subject to random errors of $\approx 15-20\%$, mostly due to uncertainties in estimating O₃ and NO₂ optical depths. This is stressed because too often climatological values of ozone are used in computing AOD, and corrections for NO₂ absorption are not made. The range in total ozone varies from about 100 Dobson Units (DU) to about 400 DU during recovery of the "ozone hole" each year at SPO, and at BRW the range is from 250 DU to 500 DU over the annual cycle. NO₂ increases by a factor of six from austral spring through summer at SPO, in correlation with photochemical processes that drive the seasonal cycle of O₃. Short-term variations also occur if the polar vortex undergoes dynamical fluctuations. Tomasi and Vitale (1998) reveal another uncertainty that occurs when estimating gaseous absorption of species as a function of airmass at low solar elevation angles. Refinements in processing AOD data may be warranted to account for such biases. Errors will be reduced more significantly, however, if coincident measurements of ozone and nitrogen dioxide are used in the derivation of AOD.

4. RECENT RESULTS FROM SPECTRAL AOD MEASUREMENTS AT SPO AND BRW

4.1 Time Series Analyses

Preliminary processing of data from SPO beginning January 2000 and from BRW beginning March 2000 indicates significant contrasts in aerosol concentrations as well as spectral signatures. Time series of one-minute resolved AOD at four wavelengths are now being generated. From the spectral data, it is possible to determine the relative size of particles as well. Relative size is inferred by evaluating a parameter referred to as the Ångström exponent a defined by a power law in which AOD $\propto \lambda^{-a}$. Specifically, a relates to the negative slope of spectral AOD when plotted on a log scale (e.g., Section

4.2, Figure 5), with larger values indicative of smaller particles. In the following analysis, values of a are evaluated for wavelength pairs (λ_1 , λ_2) using the relationship

$$\mathring{a}(\lambda_1, \lambda_2) = -\log l0(\tau(\lambda_1) / \tau(\lambda_2)) / \log l0(\lambda_1 / \lambda_2)$$
⁽²⁾

where $\lambda_1 < \lambda_2$, and $\tau(\lambda_1)$ and $\tau(\lambda_2)$ are the AODs at the respective wavelengths.

Figure 4 shows time series of spectral AOD and derived Ångström exponents during March to April 2001 at BRW. In this case, values of a were derived for the channel pairs (λ_1/λ_2) as labeled. Note that a is inversely proportional to mean particle size and tends to have lower values when AOD increases. Similar time series from SPO (not shown) reveal less variability, optical depths an order of magnitude lower (0.015 to 0.025 at 500 nm), and Ångström exponents that are larger (1.2-1.8) than at BRW indicating a dominance of smaller particles in the distribution.



Figure 4. (a) Time series of one-minute spectral aerosol optical depth derived from measurements made using the NOAA/CMDL SP02-1022, 4-channel sunphotometer at Barrow, Alaska during March-May 2001, and (b) Ångström exponents calculated for the channel pairs indicated in the legend. Means and (standard deviations) of each variable are given in the legends; (a) for respective AODs, and (b) for $d(\lambda_1, \lambda_2)$.

The fact that values of $a(\lambda_1, \lambda_2)$ for different wavelength combinations tend to approximately the same mean value suggests that the power law from which the exponents are derived is a fair representation of the spectral behavior of Arctic aerosols. However, this is not always the case, especially if the particle size distribution has multiple modes. For example, aged volcanic aerosols are typically bimodal, having a large-particle mode and a small-particle mode of higher concentration (Stone et al., 1993). If values of a are derived using regression analyses over the entire spectral range of observation, as is often done, the occurrence of multiple modes cannot be resolved. On the other hand, using (2) to determine a is also prone to error because one or both of the AOD values used in the calculation may be in error as mentioned earlier (*Section 3.3*). This is addressed further in the following section.

4.2 Bi-polar Aerosol Characterizations

The great differences observed in polar records of AOD highlights the importance of assimilating similar data from other sites to better characterize aerosols spatially and temporally. **Figure 5** and **Figure 6** illustrate the potential value of assimilating data from various sites to gain a perspective on how aerosols differ from pole to pole. Using ancillary data, primarily trajectory analyses (Harris and Kahl, 1994), source regions of different aerosols can be identified and different species can be classified according to their spectral signatures.

In Figure 5 values of spectral AOD are plotted for several aerosol types. These are referenced to an analysis made for a thin cirrus cloud observed over BRW. Values vary by an order of magnitude and certain types show spectral behavior that deviates from the assumed Ångström power law. Note also that the negative slopes of the respective curves (from the bottom, up) tend to decrease in magnitude as AOD increases, suggesting that the presence of larger particles leads to enhanced extinction.

Figure 6 shows this more clearly. Here, an attempt has been made to classify the aerosol types presented in Figure 5 by plotting values of Ångström exponent for the channel pair 412/675 nm as a function of the 500 nm AOD, AOD(500). The visible range is selected as being the most important in terms of direct radiative forcing. Interestingly, there is a nonlinear relationship between relative size and the magnitude of the optical depth, noting again that $a(\lambda_1, \lambda_2)$ is inversely proportional to particle size. Also, aerosol types tend to cluster along a best fit of the data, indicated by the dashed regression curve in the figure.



bi-POLAR AEROSOL COMPARISONS

Figure 5. Mean aerosol optical depth as a function of wavelength for different aerosol types compared with that of a thin cirrus cloud. Data were collected at the CMDL Barrow Observatory, with exception of curves labeled South Pole, TNB. NYA. and that represent conditions at South Pole and Terra Nova Bay during January, and typical spring and hazy conditions at Ny Alesund, Norway, respectively (data from TNB and NYA provided by V. Vitale and A. Herber, respectively).

Cirrus clouds are composed of large ice crystals and thus have small å values. They are optically variable because of their tenuous structure, however, so the cluster is drawn out over a range of optical depths. In this example, Asian dust appears to be thicker optically than Arctic haze and composed of larger particles. Both show non-linear behavior, with greater optical depths being associated with larger particle sizes. This functionality may be related to hygroscopic particle growth that varies with relative humidity. There is observational evidence that Ångström exponents decrease in value as particles grow hygroscopically (Carrico et al., 1998).





Figure 6. Plot of Ångström exponents, a(412/675), as a function of 500 nm optical depth. AOD(500). aerosol Clusters of points correspond to oneminute data used to determine the mean spectral AODs shown in Figure 5, with exception of points labeled TNB'00, NYAspr, and NYAhaze for which highresolution data were not available. The dashed curve is a best fit of all the data, showing that AOD(500) increases for decreasing values of a(412/675), or as particle size increases.

Under pristine conditions, represented by the SPO January cluster (Spole) and the summertime background for BRW (SumBG), size spectra appear highly variable but have a very narrow range of optical depths. It is possible that this feature is an artifact of measurement because the analyses are based on data collected near the lower limit of the photometers' sensitivity. Also, because NO₂ and O₃ absorb radiation at 412 nm and 675 nm, respectively, depending on their concentrations, estimates of a(412/675) are subject to greater uncertainty when values of AOD are small because column amounts of these gases are not always measured accurately (e.g., Section 3.3). The cluster means are probably valid, however, and show that clean polar air contains a mix of very small particles. Average summer conditions at Terra Nova Bay and during spring at Ny Alesund are similar to those during spring at BRW (SprBG) as might be expected. All three sites are at sea level, at similar latitudes, and are influenced directly by marine aerosols. In contrast, SPO during summer is an extremely clean site because it is high on the Plateau and far from sources of sea salt aerosols. Occasionally during summer, the Barrow atmosphere is nearly as clear optically as at South Pole but in general, the Arctic is not as pristine a region as is often thought. Turbid conditions often perturb the radiation balance of the region as evidenced by the signatures of 'haze" and "dust" shown in Figure 6.

The analysis summarized in Figure 6 is intriguing and suggests that with a much larger statistical ensemble of observations, a bi-polar characterization of aerosols (and possibly thin clouds) could be synthesized. Taking this a step further, utilizing coincident

in situ data to determine chemical and physical properties of the particles, should provide a basis for parameterizing aerosols (and thin clouds) in climate models. Alternatively, using an inversion scheme (e.g., King et al., 1978) to infer size spectra, and Mie theory to calculate optical properties of these spectra, it may be possible to develop representative microphysical models, defined by the key radiative properties that are needed to estimate radiative forcing. Ogren (1995) lists the properties of aerosols that are required for climate studies and also stresses the need for reliable, consistent statistical representations of aerosols for such studies. Clearly, a sustained effort to obtain continuous measurements of spectral AOD at a number of high latitude sites is warranted. These will augment efforts already being made under the sponsorship of the AErosol RObotic NETwork (AERONET) (Holben et al. 2001); http://aeronet.gsfc.nasa.gov. As of this writing, no AERONET site is currently operational north or south of about 55° of latitude. Unfortunately, satellite-derived aerosol properties are limited to oceanic regions and are not available for high latitudes either. IPCC (2001) recognizes the limitations of satellite-derived properties of aerosols, stressing the need for "systematic ground-based measurements" including "total column properties such as aerosol optical depth, Angstrom coefficient..." etc.

5. SUMMARY AND FINAL REMARKS

The polar regions play an important role in global change studies because *enhanced warming* is predicted for high latitudes as greenhouse gas concentrations continue to increase in the atmosphere. Initial warming may cause a positive feedback due to diminishing snow and ice cover that lowers the planetary albedo. This process is poorly understood and not well simulated by climate models partly because the radiative effects of clouds and aerosols are inadequately parameterized. A very preliminary characterization of polar aerosols has been presented to show how variable they are spatially and temporally. It is clear that a much better understanding of polar aerosols is needed. Through international collaboration an improved understanding and characterization of polar aerosols and their radiative effects can be realized.

5.1 Current Status of Polar Aerosol Studies

The analysis presented in Figures 5 and 6, though based on a limited data set, demonstrates the value in using spectral AOD data for characterizing diverse species of aerosols that impact polar radiation budgets. Ultimately, it is the radiative forcing by aerosols that is of primary concern to climatologists. Theoretical calculations suggest that the polar regions are peculiar in this regard. Cacciari et al. (2000), for instance, show that aerosol radiative forcing can change sign depending on chemical species, surface properties and solar geometry. Particle size is another critical property that varies hygroscopically with relative humidity. Unfortunately, surface measurements may be inadequate for high-latitude aerosol studies because polar atmospheres are highly stratified, mixing is suppressed, and aerosols often reside in layers above the surface-based temperature inversion. Only by combining (columnar) AOD data with in situ surface and airborne observations can advancements in our understanding of aerosol radiative forcing be made. Such investigations must be correlated with dynamical

analyses and cloud observations if the indirect effects of aerosols are to be determined. Even slight changes in cloud microphysical or physical properties resulting from interactions with aerosols are likely to perturb the climate of the high latitude regions because clouds profoundly impact the radiation balance there (Stone, 1997; Stone, 1993; Stone and Kahl, 1991). It is in recognition of these uncertainties that SCAR and IPCC recommend upgrading observational networks, to include sustained measurements of aerosol optical properties in polar regions. Unfortunately, AERONET (Holben et al., 2001) has not yet deployed any of its standard photometers to polar sites, and satellite data are not yet useful at high latitudes for deriving aerosol properties. Therefore, alternative steps are recommended to meet the scientific goal of producing a *global* aerosol climatology.

5.2 Proposed International Polar AOD Network and Archive

Steps have been taken by researchers from several countries to coordinate their activities in this regard. Interested countries include Italy, Australia, Germany, Japan, the United States, Russia, the United Kingdom, and France. AOD observations are currently being made at eight sites, six in Antarctica (Terra Nova Bay, Halley Bay, Neumayer, Syowa, Davis, and South Pole) and two in the Arctic (Barrow, Alaska and Ny Alesund (at Koldewey), Norway). Historically, data are also available from Russian Antarctic stations and possibly from Siberia. Although a variety of photometers have been used and the data records are of variable length and continuity, there is agreement that an archive of polar AOD data, including metadata that describes field sites, calibration procedures, and operational activities, will provide the basis for better spatial and temporal characterizations of aerosols. Initially, efforts should focus on establishing "background" values from which perturbations, whether from volcanic activity, anthropogenic or other sources, can be referenced. As evidenced in Figure 1, the high latitudes are currently experiencing a period of volcanic quiescence; i.e., no major eruptions in recent years have affected BRW and SPO, and AOD values are currently below historical minima. Therefore, the opportunity to establish seasonal cycles of AOD, and derived properties on a bi-polar basis now exists. Once these cycles are understood, analyses in conjunction with model studies should provide the means to more accurately quantify the radiative effects of aerosols at high latitudes.

The Italian Program for Antarctic Research (PNRA) has taken a lead in polar AOD investigations by making very precise spectral measurements at the Italian base, Terra Nova Bay (e.g., Vitale and Tomasi, 1990; Tomasi, et al., 1992). It is hoped that with further support PNRA will coordinate international efforts to assure that consistent, high quality data from the network be assimilated and be made available to the scientific community. Having a network essentially in place, the following activities are recommended:

- Convene an international workshop for presentations of preliminary results from the various polar programs, discuss common strategies, future needs/goals, protocol, logistics of intercalibration activities, data format, etc.
- Establish an archive of polar AOD data and metadata

- Establish a website to give easy access to these data, bibliographical materials, with links to related sites
- Provide a foundation for international collaboration to further our understanding of polar aerosols utilizing observational, as well as theoretical methodologies

5.3 The Role of CMDL pertaining to the International Polar AOD Network and Archive

The Baseline Surface Radiation Network (BSRN) (Ohmura et al., 1998) has been proposed as a model of how to establish the kind of archive mentioned above. Certain operational procedures and calibration practices of AERONET should also be considered. CMDL is an active participant in BSRN, operating five of the existing 18 sites in that network, including BRW and SPO. Recent enhancements of the existing radiation monitoring programs at SPO and BRW, to include continuous photometric observations, have been successful. The CMDL Radiation Group will endeavor to sustain these projects and cooperate with the international community. The goal will be to provide high quality AOD data from SPO and BRW for a myriad of climate studies. Evaluations of the direct and indirect radiative forcing by polar aerosols will be undertaken in conjunction with ongoing measurements of net surface radiation made at the observatories (Stone, 2000; Stone et al., 1996; Dutton et al., 1989). These will be correlated with in situ data derived from nephelometers, LIDAR data when available, and ancillary data to determine the source and evolution of aerosols observed. These investigations will contribute to improving parameterizations of aerosol forcing in climate models, and permit more accurate interpretations of remote sensing data that, to varying degrees, are contaminated by the intervening aerosol layers.

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References

- Barrie, L.A., Arctic air pollution, An overview of current knowledge, *Atmos. Envioron.*, 20, 643-663, 1986.
- Bergin, M.H., E.A. Meyerson, J.E. Dibb, and P.A. Mayewski, Relationship between continuous aerosol measurements and firn core chemistry over a 10-year period at the South Pole, *Geophys. Res. Let.*, 25(8), 1189-1192, 1998.
- Bodhaine, B.A., Central Antarctica, atmospheric chemical composition and atmospheric transport. Chemical Exchange Between the Atmosphere and Polar Snow, E.W. Wolff and R.C. Bales (eds.), *NATO ASI Series I, 43:145-172*, Springer-Verlag, Berlin, 1996.

- Bodhaine, B.A., The U.S. aerosol monitoring program in Antarctica, Proceedings, 4th Workshop Italian Research on Antarctic Atmosphere, Porano IV, Italy, October 21-23, 1991, Italian Physical Society, Bologna, Italy, 15-25, 1992.
- Bodhaine, B.A., N.B. Wood, E.G. Dutton, and J.R. Slusser, On Rayleigh optical depth calculations, *Jou. Atmos. and Ocean. Tech.* 16, 1854-1861, 1999.
- Bodhaine, B.A., and E.G. Dutton, A long-term decrease in arctic haze at Barrow, Alaska. *Geophys. Res. Lett.*, 20, 947-950, 1993.
- Cacciari, A., A. Lupi, C. Tomasi, V. Vitale, and S. Marani, Calculation of the radiative forcing caused by aerosol particles in Antarctic regions (Terra Nova Bay), 8th Workshop Italian Research on Antarctic Atmosphere, October 20-22, 1999, Italian Physical Society, Bologna, Italy, 455-467, 2000.
- Carrico, C.M., Rood, M.J., Ogren, J.A., Aerosol light scattering properties at Cape Grim, Tasmania, during the First Aerosol Characterization Experiment (ACE 1), J. Geophys. Res., 103, 16565-16574, 1998.
- Charlson, R.J., S.E. Schwartz, J.M. Hales, R.D. Cess, J.A. Coakley, J.E., Hansen, and D.J. Hofmann, Climate forcing by anthropogenic aerosols, *Science*, *255*, 423-420, 1992.
- Dutton, E.G., and J.R. Christy, Solar radiative forcing at selected locations and evidence for global lower tropospheric cooling following the eruptions of El Chichon and Pinatubo, *Geophys, Res. Lett.*, 19(23), 2313-2316, 1992.
- Dutton, E.G., R.S. Stone, and J.J. DeLuisi, South Pole radiation balance measurements April 1986 to February 1988, NOAA Data Report ERL ARL-17, Boulder, CO, 49 pp., 1989.
- Harris, J.M., and J.D.W. Kahl, Analysis of 10-day isentropic flow patterns for Barrow, Alaska; 1985-1992, J. Geophys. Res., 99(D12), 25,845-25,855, 1994.
- Hofmann, D.J., and J.M. Rosen, On the prolonged lifetime of the El Chichon sulfuric acid aerosol cloud, J. Geophys. Res., 92(D8), 9825-9830, 1987.
- Holben, B.N., et al., An emerging ground-based aerosol climatology: Aerosol Optical Depth from AERONET, J. Geophys. Res., 106, 12 067-12 097, 2001.
- IPCC, 2001, Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, [Houghton, J.T. et al., [eds.)]. Cambridge Univ. Press, Cambridge, United Kingdom and New York, NY, USA, 881pp, 2001.
- Jaffe, D., T. Iversen, and G. Shaw, Comment on "A long term decrease in arctic haze at Barrow, Alaska" by B.A. Bodhaine and E.G. Dutton, *Geophys, Res. Lett., 22(6),* 739-740, 1995.
- King, M.D., D.M. Byrne, B.M. Herman, and J.A. Reagan, Aerosol size distributions obtained by inversion of spectral optical depth measurements, *J. Atmos. Sci.*, 35, 2153-2167, 1978.
- Ogren, J.A. A systematic approach to in situ observations of aerosol properties, *Aerosol forcing of climate*, R.J. Charlson and J. Heintzenberg (eds.), Wiley & Sons, New York, 215-226, 1995.
- Ohmura, A., *et al.*, Baseline surface radiation network (BSRN/WCRP): New precision radiometry for climate research., *Bulletin of the American Meteorological Society* 79(10), 2115-2136, 1998.
- Reddy, P.J., *et al.*, Aerosol optical depths over the Atlantic derived from shipboard sunphotometer observations during the 1988 global change expedition, *Global Biochem. Cycles.*, 4(3), 225-240, 1990.
- Quinn, P.K., T.L. Miller, T.S. Bates, J.A. Ogren, E. Andrews, and G.E. Shaw, A Three-Year record of simultaneously measured aerosol chemical and optical properties at Barrow, Alaska, J. Geophys. Res., (in press), 2002.

Shaw, G.E., Sun photometry, Bull. Am. Meteorol. Soc., 64(1), 4-10, 1983.

Shaw, G.E., and M.A. Khalil, Arctic haze, in *The Handbook of Environmental Chemistry, Vol.* 4/Part B, Hutzinger, Ed., Springer-Verlag, 69-111, 1989.

- Schnell R.C., D.B. King, and R.M. Rosson (eds.), *Climate Monitoring and Diagnostics Laboratory* Summary Report No. 25, 1998-1999, Boulder, CO, 154 pp., 2001.
- Stone, R.S., Climate monitoring at Barrow, Alaska and South Pole: An overview of U.S. studies of polar surface radiation balance and aerosols, Proceedings of the 8th Workshop Italian Research on Antarctic Atmosphere, Bologna, Italy, October 20-22, 1999, Italian Physical Society, Bologna, Italy, eds. M. Colacino and G. Giovanelli, 83-98, 2000.
- Stone, R.S., Variations in Western Arctic temperatures in response to cloud radiative and synopticscale influences, J. Geophys. Res., 102(D18), 21,769-21,776, 1997.
- Stone, R.S., Properties of austral winter clouds derived from radiometric profiles at South Pole, J. Geophys. Res., 98(D7), 12,961-12,971, 1993.
- Stone, R.S., E.G. Dutton, J.M. Harris, and D. Longenecker, Earlier Spring Snowmelt in Northern Alaska as an Indicator of Climate Change, *J. Geophys. Res.*, (in press), 2002.
- Stone, R., T. Mefford, E. Dutton, D. Longenecker, B. Halter, and D. Endres, Barrow, Alaska, Surface radiation and meteorological measurements: January 1992 to December 1994, NOAA Data Rep. ERL CMDL-11, Boulder, CO, 81pp., 1996.
- Stone, R. S., E.G. Dutton, and J.R. Key, Properties and Decay of Pinatubo Aerosols in Polar Regions Compared with Tropical Observations, *Eighth Conference on Atmospheric Radiation, Nashville, Tenn.*, Jan. 23-28, 1994, American Meteorological Society, 432-434, 1994.
- Stone, R.S., J. Key, and E. Dutton, Properties and decay of stratospheric aerosols in the Arctic following the 1991 eruptions of Mount Pinatubo, *Geophys. Res. Let.*, 20(21), 2359-2362, 1993.
- Stortini, A.M., R. Cini, R. Traversi, R. Udisti, and G. Piccardi, Surface active substance effect on marine aerosol fractionation as function of altitude, 8th Workshop Italian Research on Antarctic Atmosphere, October 20-22, 1999, Italian Physical Society, Bologna, Italy, 411-420, 2000.
- Tomasi, C., and V. Vitale, Calculation of the relative optical mass functions for air, water vapour, ozone and nitrogen dioxide in the Antarctic and Arctic Atmospheres, 7th *Workshop Italian Research on Antarctic Atmosphere*, Conference Proceedings Vol. 62, October 22-24, 1997, Italian Physical Society, Bologna, Italy, 253-282, 1998.
- Tomasi, C., V. Vitale, G. Zibordi, Multiwavelength sun-photometric measurements of the atmospheric turbidity parameters at Terra Nova Bay during January 1990, 3rd Workshop Italian Research on Antarctic Atmosphere, Porano III, October 22-24, 1990, Italian Physical Society, Bologna, Italy, 125-142, 1992.
- Traversi, R., S. Becagli, E. Castellano, O. Largiuni, and R. Udisti, Stability of chemical species in firm layers from Antarctica, 8th Workshop Italian Research on Antarctic Atmosphere, October 20-22, 1999, Italian Physical Society, Bologna, Italy, 421-443, 2000.
- Vitale, V., and C. Tomasi, Atmospheric turbidity measurements at Terra Nova Bay with the multispectral sun-photometer model UVISIR", 2nd Workshop Italian Research on Antarctic Atmosphere, Porano II, October 19-20, 1989, Italian Physical Society, Bologna, Italy, 89-104, 1990.