An Empirical Approach to Aerosol Model Development for Radiative Transfer Calculations

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Introduction

Standard aerosol models are typically used to represent aerosol optical and microphysical properties for radiative transfer modeling. While surface measurements of aerosol properties are available, these data may not be used because the required parameters are not directly measured and extraction can be time consuming. Empirically derived aerosol models from two Atmospheric Radiation Measurement (ARM) Program sites are presented here to assess the similarity of the most commonly used standard aerosol models to observations in these particular regions, and to provide a more representative set of aerosol optical properties for modeling radiative fluxes at these sites. The data-driven models can be used to illustrate the amount of aerosol-induced uncertainty in radiative transfer calculations based on the specification of generalized aerosol types rather than measurements and can be useful in quantifying the extent of closure between measurements and model calculations. They are also intended to provide a reasonable set of model inputs for determining aerosol impacts at these sites.

Long-term observations (1998 to 2002) made by National Oceanic and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory (CMDL) have been used to classify aerosol types from two ARM sites: Southern Great Plains (SGP) at Lamont, Oklahoma (mid-latitude continental) and North Slope of Alaska (NSA) at Barrow (BRW), Alaska (Arctic marine). Distinct aerosol types have been extracted from these data sets using cluster analysis with in situ observations of aerosol chemistry, optical properties, and local meteorology. The fundamental aerosol types at each site are presented as well as a time series and the range of values for properties relevant to radiative transfer modeling. Information from isentropic air trajectory analysis from each of the sites is used to further characterize the different aerosol types. Finally, a comparison is given between these results and a standard model.

Cluster Analysis

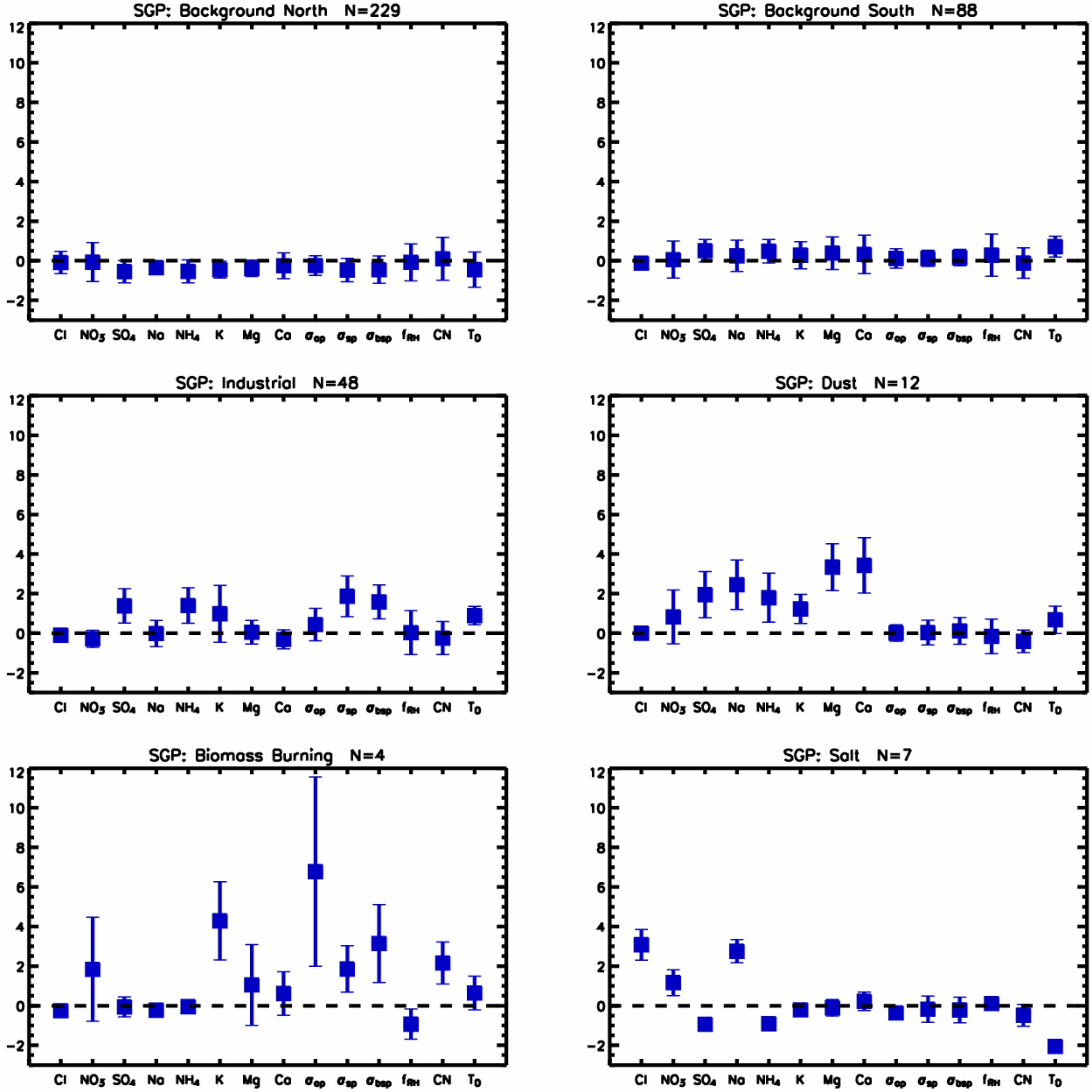
A k-means clustering algorithm was used with daily averaged aerosol properties to identify aerosol types based on patterns inherent in the data. All data used in this analysis is for aerosol with particle diameter (D_p) less than 1 μm. Variables included in the analysis are those that are directly measured, rather than derived, and most strongly differentiate aerosol types. These include chemical composition (cation and anion concentrations); absorption (σ_{ap}), scattering (σ_{sp}) and backscatter (σ_{bsp}) coefficients; aerosol number concentration (CN); and dew point temperature (T_d). All aerosol optical properties are measured at a wavelength of 550 nm. Chemical concentrations, essential to the well defined clusters, are available at SGP on a daily basis for the $D_p < 1$ μm and approximately every three days at Barrow. While aerosol optical properties are available at a higher frequency, the chemistry data dictates the temporal resolution of the data set used here.

Chemical components measured at BRW vary slightly from SGP with the addition of methanesulfonate (MSA-), sea salt (SS), and non-sea-salt SO_4=, K+, Mg+2, and Ca+2 (identified with *). The dependence of aerosol light scattering with changes in relative humidity, f(RH), is included in the SGP analysis but f(RH) measurements are not made at BRW. Here, f(RH) is the ratio of light scattering at 85% RH to 40% RH. Each box in Figures 1a and 1b represents one of six different clusters for each site. All data were standardized prior to the analysis (the time series of each variable were transformed to mean of zero and variance of one) so that values for all variables are relative. Boxes indicate the mean loading of the variable within each cluster and the bars represent the variance (one standard deviation). Clusters have been subsequently named to represent aerosol type.

The analysis requires a valid value for each variable so any day with missing data for any of the chosen variables was excluded leaving 388 days of observations at SGP and 360 days at BRW. The number of members for each cluster are shown next to the cluster name. The most commonly occurring aerosol types show variable loadings close to mean values (zero) while rare or ’event driven’ aerosols typically have high loadings for a few definitive variables. For example, SGP has two very similar aerosol types that are present most of the year while other types are observed on only a few days. At Barrow there are also two common aerosol types, ocean biogenic and Arctic haze, that are seasonally dependent. The other aerosol types at Barrow occur much less frequently.

Aerosol Properties

The range of values for extinction coefficient (σ_{ext}), single scattering albedo (ω_0), asymmetry factor (g), and Ångström exponent (å) for each aerosol type are presented as box-and-whisker plots in Figures 2a and 2b. These variables are defined in Table 1. The middle line of the box represents the median value, the top and bottom of the box the 75th and 25th percentile values, and the top and bottom of the whiskers the 95th and 5th percentile values. Values are absolute, i.e., they are not standardized. The first three properties are generally used in radiative transfer computations to define the aerosol. The six aerosol types at each site are well differentiated by these properties indicating that a type may be identified in near-real time based on a small suite of observations. The median values also provide an estimate of reasonable optical properties for inputs into radiative transfer models.


SGP: Background North N=229
SGP: Background South N=88
SGP: Industrial N=48
SGP: Dust N=12
SGP: Biomass Burning N=4
SGP: Salt N=7
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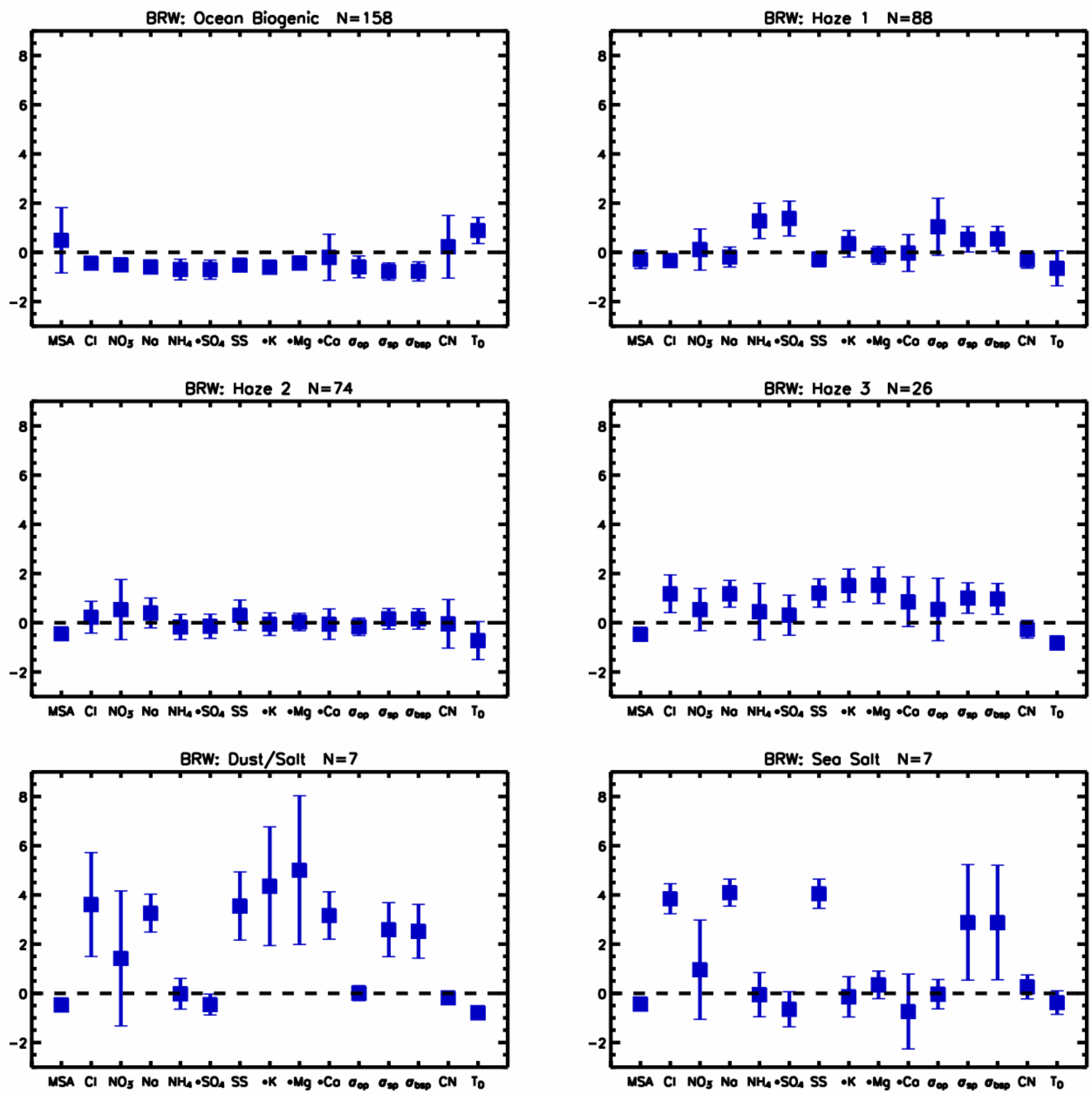
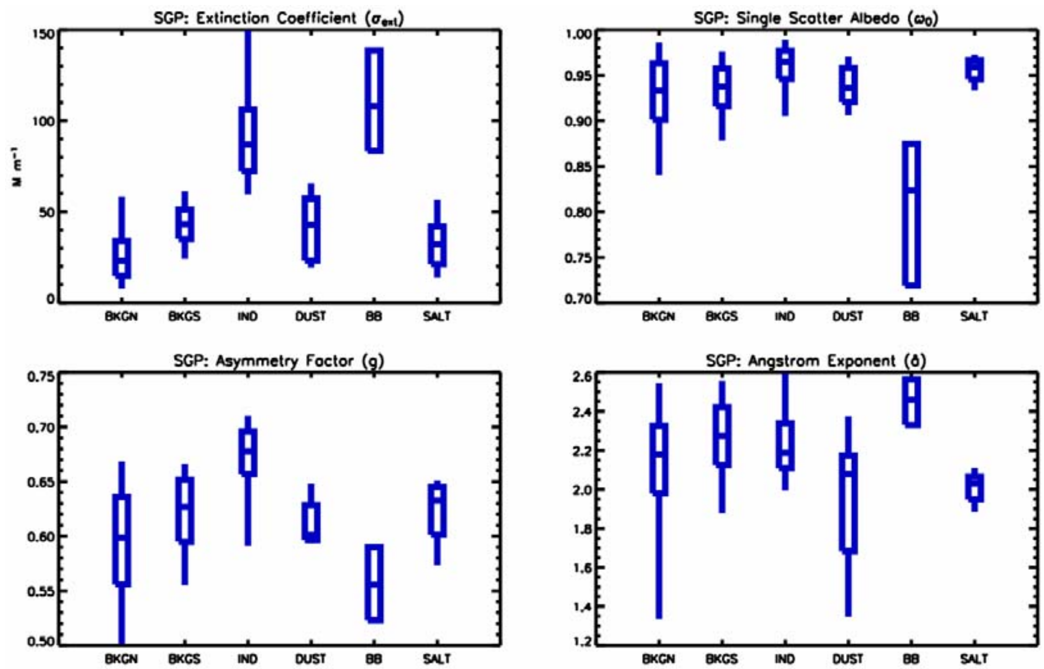
Cl NO3 SO4 Na NH4 K Mg Ca σap σsp σbsp fRH CN To

(a)

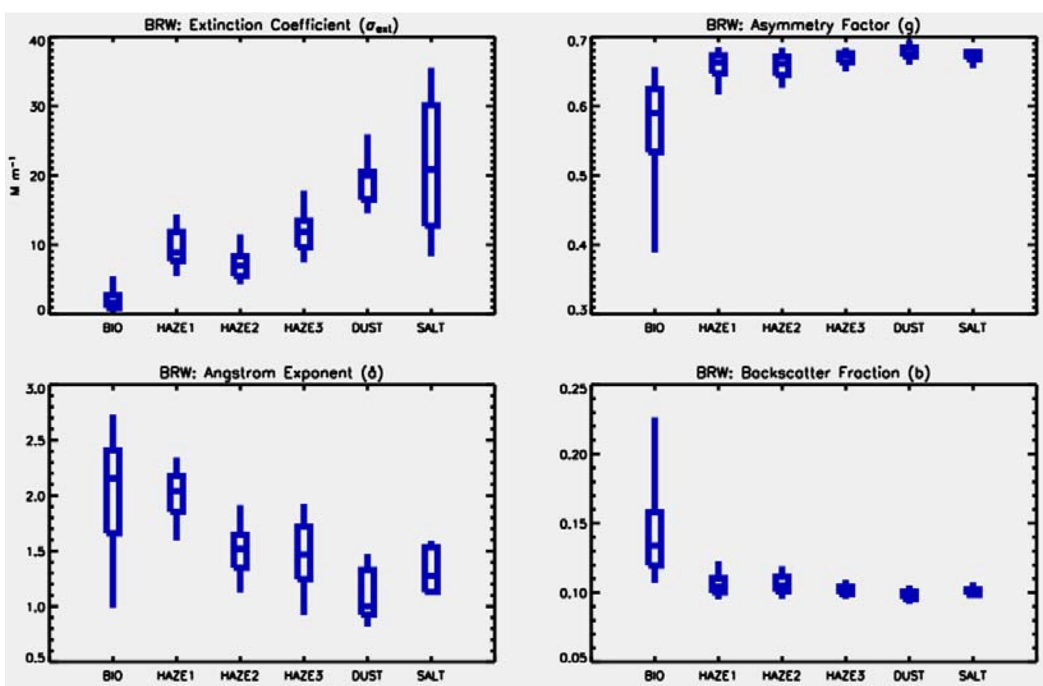


(b)

Figure 1. (a) Results of cluster analysis for SGP. Variables are standardized to a mean of zero and variance of one. (b) Results of cluster analysis for Barrow. Variables are standardized to a mean of zero and variance of one.



(a)



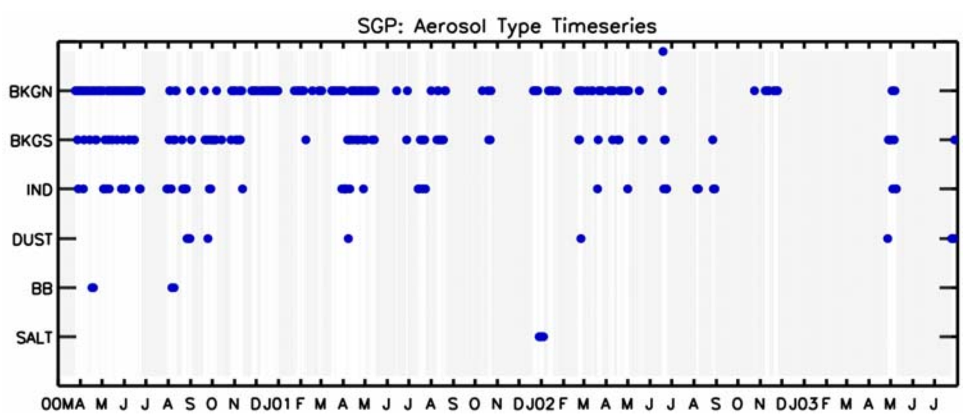
(b)

Figure 2. (a) Range of values for selected optical properties for the six SGP aerosol types. (b) Range of values for selected optical properties for the six Barrow aerosol types.

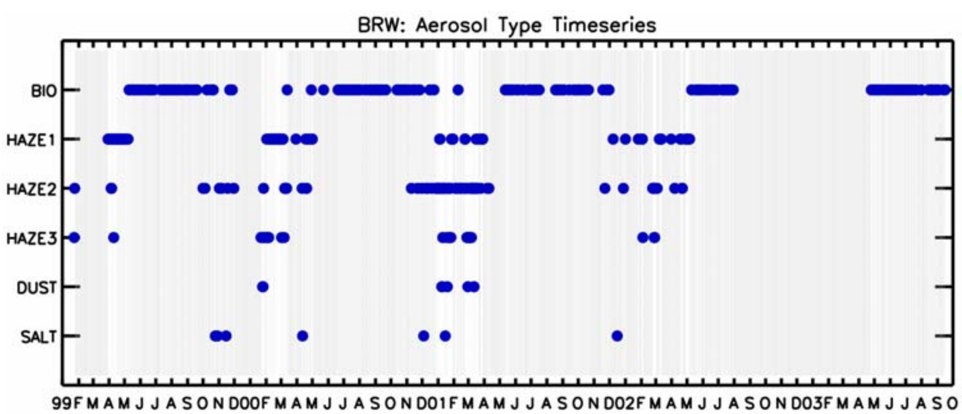
Table 1. Aerosol properties relevant to radiative transfer modeling.	
$\sigma_{ext} = \sigma_{sp} + \sigma_{ap}$	Scattering plus absorption.
$\omega_0 = \sigma_{sp} / (\sigma_{sp} + \sigma_{ap})$	Ratio of scattering to extinction.
$g = 0.95 - 2.9979b + 2.3059b^2$	A function of *b*, backscatter fraction, from Wiscombe and Grams, 1976.
å = $-\log[\sigma_{sp}(550nm)/\sigma_{sp}(700nm)]/\log[550nm/700nm]$	Green to red wavelength.

Time Series and Trajectory Analysis

Cluster or aerosol type membership for daily averaged observations are plotted in a time series (Figures 3a and 3b) to illustrate any seasonal or temporal trends in the occurrence of aerosol types. In the timeseries plots, dots represent which cluster a particular day's observation belongs to and the grey



(a)



(b)

Figure 3. (a) Timeseries of SGP aerosol types. Dots represent daily averaged observations and grey lines represent missing daily averaged data. (b) Timeseries of Barrow aerosol types. Dots represent daily averaged observations and grey lines represent missing daily averaged data.

lines represent missing data. In general, aerosol types follow seasonal patterns that are loosely related to source region or trajectory. Five day back-trajectories clusters (Polissar et al. 2001) arriving at the sites at a height of 500 m have been computed along with their percent occurrence for each aerosol type. At SGP, the three most common aerosol types are related to industry (based on the chemical composition), one corresponding to colder air masses and more northerly trajectories and the others to warmer air masses and a southern origin. The composition and optical properties of these aerosol types differ, although they would all be considered standard 'urban' aerosols. At Barrow, as demonstrated in Quinn et al. 2002, ocean biogenic (MSA) aerosol dominates in the summer when open ocean prevails and Arctic Haze dominates in the winter and early spring. Some aerosol is ubiquitous (e.g., Arctic Haze) at particular times of the year and seems to correspond to most trajectories. For this reason, back-trajectory analysis used by itself may not be the best indicator of aerosol type.

Input Quantities for Aerosol Radiative Impact Studies: Comparison of Empirical and Theoretical Aerosol Models

The above analysis identifies and defines aerosol types at two ARM sites and shows the range in optical properties relevant to radiative transfer modeling for each type. Mean values for the key quantities necessary for computations are summarized below. To understand how the new empirical models compare to the standard, theoretical models commonly used in radiative transfer, comparisons of some key variables are given. Reported quantities include the mass extinction efficiency (Q_e) an expression of the radiative impact of an aerosol per unit mass, the single scattering albedo which indicates the fraction of total extinction due to scattering (ω_0) versus absorption ($1-\omega_0$), and the asymmetry factor (g) which indicates directional scattering. The standard model presented here is that of Shettle and Fenn (1979), commonly used in radiative transfer models. In this model, four different aerosol types are provided (indicated under 'MODEL' in the table below) which have been matched most closely to the empirical aerosol models derived here from the ARM sites (indicated under 'DATA' below). Standard models provide an extinction coefficient based on aerosol number density which can not be directly compared to the mass extinction efficiency presented here. For comparison purposes, the average optical depth (tau) for each aerosol type has been computed from the extinction coefficients and extinction efficiencies at average ambient relative humidity using meteorological visibility and a standard vertical aerosol distribution model (McClatchey 1972), as in SBDART (Ricchiazzi et al. 1998). In this case, the differences in optical depth are due only to the differences in the extinction parameter defined by the models and observations. Data for SGP only is shown here as the optical depth calculation requires an f(RH) value not available for BRW.

Table 2. Mean aerosol optical properties for the newly derived aerosol types as compared to optical properties from the standard model of Shettle and Fenn (1979).

	Data bkgn	Model urban	Data bkgs	Model urban	Data ind	Model urban	Data dust	Model rural	Data BB	Model --	Data salt	Model maritime
Q_e	4.42	--	3.63	--	6.11	--	1.61	--	5.16	--	9.50	--
tau	0.051	0.107	0.098	0.093	0.182	0.169	0.074	0.106	0.202	--	0.069	0.102
ω_0	0.933	0.703	0.938	0.726	0.965	0.726	0.936	0.943	0.824	--	0.959	0.987
g	0.60	0.700	0.63	0.71	0.68	0.71	0.60	0.653	0.56	--	0.63	0.718

Summary of SGP Aerosol Types

- The SGP site is dominated by a low level of industrial aerosol year round.
- The chemical composition of industrial aerosol varies with season and source, dictated by northerly versus southerly trajectories. However, most of the optical properties between the different industrial aerosol types are not distinctly different.
- The steady aerosol climate is punctuated by events such as high winds, fire, and winter storms that bring aerosols with distinctly different optical properties.

Summary of Barrow Aerosol Types

- The BRW site is dominated by biogenic aerosol of oceanic origin in the summer and haze in the winter.
- Haze aerosol consists of various chemical components that prevail at different times and have distinct optical properties.
- Long-range transport brings dust and sea salt to the region primarily in the wintertime.

Conclusions and Future Directions

- At both sites, the range of optical and microphysical properties for each of the aerosol types are significantly distinct to be used with clustering algorithms to define empirically driven aerosol models.
- Mean values of key aerosol properties have been defined that can be used as inputs for radiative transfer modeling at the ARM SGP and BRW sites.
- Mean optical properties of the model agree well for some aerosol types for which there is a good 'match'. However, variability in the actual conditions at SGP indicate that a standard model may introduce error into radiative impact studies.
- A modeling study will be conducted to determine the radiative impacts of the different aerosol types at each site using the derived inputs and how they differ from the standard models.
- A discriminant analysis, or reverse cluster analysis, will be used to develop a classification algorithm to identify an aerosol type based on routinely measured properties. In this way, daily observations can be used to determine appropriate optical properties for use in model computations.

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References

McClatchey, R. A., R. W. Fenn, J.E.A. Selby, F. E. Volz, J. S. Garing, 1972: Optical Properties of the Atmosphere. AFCRL-72-0497 Environmental Research Papers No. 411.

Polissar, A. V., P. K. Hopke, and J. M. Harris, (Need date): Source Regions for Atmospheric Aerosol Measured at Barrow, Alaska. *Environmental Science & Technology*, **35**(21), 4214-4226.

Quinn, P. K., T. L. Miller, T. S. Bates, J. A. Ogren, E. Andrews, and G. E. Shaw, 2002: A 3-year record of simultaneously measured aerosol chemical and optical properties at Barrow, Alaska. *Journal of Geophysical Research*, **107**, 10.1029/2001JD001248.

Ricchiazzi, P., S. Yang, C. Gautier, and D. Sowle, (Need date): SBDART: A Research and Teaching Software Tool for Plane-Parallel Radiative Transfer in the Earth's Atmosphere. *Bulletin of the American Meteorology Society*, **79**(10), 2101-2114.

Shettle, E. P., and R. W. Fenn, 1979: Models for the Aerosols for the Lower Atmosphere and the Effects of Humidity Variations on Their Optical Properties. AFGL-TR-79-0214 Environmental Research Papers No. 676.

Wiscombe, W. J., and G. W. Grams, 1976: The Backscattered Fraction in Two-Stream Approximations. *Atmospheric Sciences*, **33**(12), 40-2451.