Development of Aerosol Models for Radiative Flux Calculations at ARM Sites: Utility of Trajectory Clustering for Characterizing Aerosol Climatology

E. Andrews
Cooperative Institute for Research in the Environment
University of Colorado
Boulder, Colorado

E. Andrews, J. A. Ogren, P. J. Sheridan, and J. M. Harris
Climate Monitoring and Diagnostics Laboratory
National Oceanic and Atmospheric Administration
Boulder, Colorado

P. K. Quinn
Pacific Marine Environmental Laboratory
National Oceanic and Atmospheric Administration
Seattle, Washington

Abstract

The uncertainties associated with assumptions of generic aerosol properties in radiative transfer codes are unknown, which means that these uncertainties are frequently invoked when models and measurements do not agree. In general, the radiative calculations require specification of the aerosol optical depth $\delta(\lambda)$, single-scattering albedo $\varpi_o(\lambda)$, and asymmetry parameter $g(\lambda)$, all as functions of wavelength λ and altitude. Here we use existing aerosol data from the Atmospheric Radiation Measurement (ARM) Southern Great Plains (SGP), and North Slope of Alaska (NSA) sites to begin development of a set of "models" that describe the radiative properties of different types of aerosols at these sites. These models aim to provide an internally consistent specification of aerosol optical and microphysical properties, compatible with the requirements of standard radiative transfer codes. For each of these models of the different aerosol types, we use the aerosol data to determine the range of aerosol properties that are included in that type. This will allow us to estimate the aerosol-induced uncertainty in radiative flux calculations that are based on specification of aerosol types rather than on coincident aerosol measurements.

Aerosol properties at the SGP and NSA sites show considerable variability on multiple time scales (Delene and Ogren 2001). Our hypothesis is that this variability is not random, but rather is connected with changes in the types of particles prevailing at any given time. This hypothesis is supported by the observation that aerosols at both the SGP and NSA sites absorb more and backscatter more in the cleanest air, which suggests that the pollution aerosols have very different characteristics from aerosols encountered during clean periods at these sites (Delene and Ogren 2001). We anticipate that airmass

origin will be the dominant meteorological parameter influencing the aerosol type. Here, we segregate surface aerosol properties at SGP and NSA as a function of airmass back trajectory to provide a first cut at identifying aerosol types and developing aerosol models for these two sites.

The first step in aerosol model development is to determine the prevailing aerosol types at SGP and NSA. We use the trajectory cluster approach described by Harris and Kahl (1994) to identify aerosol source regions and segregate the aerosol optical data based on the trajectory clusters (see Figure 1). The input data to the statistical analyses includes aerosol light extinction, single-scattering albedo, Ångström exponent, back scattering fraction and sub-micron scattering and absorption fractions. These statistical analyses require long, homogeneous datasets, so we have chosen a subset of the aerosol data for which the longest data set is available (6 years at SGP, 5 years at NSA). However, the sites have other aerosol data available for a shorter time period (e.g., aerosol chemistry, hygroscopic growth, and aerosol optical depth). The aerosol types are derived from the statistical analysis of the long-term measurements based on the airmass trajectories while the full suite of measurements can be used both further constrain the aerosol type (e.g., polluted marine vs. marine) and also to determine the complete set of radiative properties for each type. The shorter-term records can also be used to examine the clusters for consistency; for example, we would expect that the chemical composition would be similar for members of each cluster, and the presence in a cluster of samples with very different chemical compositions would suggest issues with the clustering.

Significant differences are evident in some (but not all) long-term aerosol optical properties for the trajectory clusters at the two sites. Trajectories arriving at the NSA site from Siberia (T2 on Figure 1a) have higher aerosol loading (high extinction) and somewhat darker (low ϖ_o) particles than for other trajectory types (Figure 2). These Siberian airmasses are temporally consistent with the springtime Arctic Haze phenomenon. In contrast, the summertime trajectories originating in the Bering Sea (T4 on Figure 1a) have the lowest aerosol loading of any of the six trajectory clusters. The chemistry data for the Bering Sea trajectory cluster are suggestive of a clean marine aerosol type, with high mass fractions of Na^+ and MSA. Both the Arctic Haze and clean marine aerosol observations are consistent with the results reported by Quinn et al. (2002) at the NSA site.

At the SGP site, the two trajectory clusters originating southwest (SW) of the site and passing over the Texas and Louisiana urban and industrial areas (T1 and T2 in Figure 1b) have the highest aerosol loading (Figure 3) and K^+ fraction (a marker for biomass burning) but can be distinguished from each other based on other chemistry and particle size measurements. T2, which extends past the Gulf of Mexico and into the Caribbean Sea has larger particles and chemistry indicative of a marine aerosol (high Na^+), while the T1 cluster which originates closer to the coast has the highest SO_4 and NH_4 mass fractions of any of the six identified trajectory clusters. In contrast to these polluted airmasses arriving at the SGP site, the three trajectory clusters originating to the northwest of the SGP site (T4, T5, and T6) have very low aerosol loadings (they have about half the aerosol extinction of the SW trajectory clusters). T6, which originates south and west of the Aleutian Islands, has the lowest ϖ_o of any of the trajectory clusters and the highest Ca^+ mass fraction. The low ϖ_o and high Ca^+ may be suggestive of intercontinental transport of aerosol (pollution+dust) from Asia, but may also indicate a very aged airmass in which the hygroscopic aerosol has been removed by cloud and precipitation scavenging. The Ca^+ may also be due to local dust during high winds.

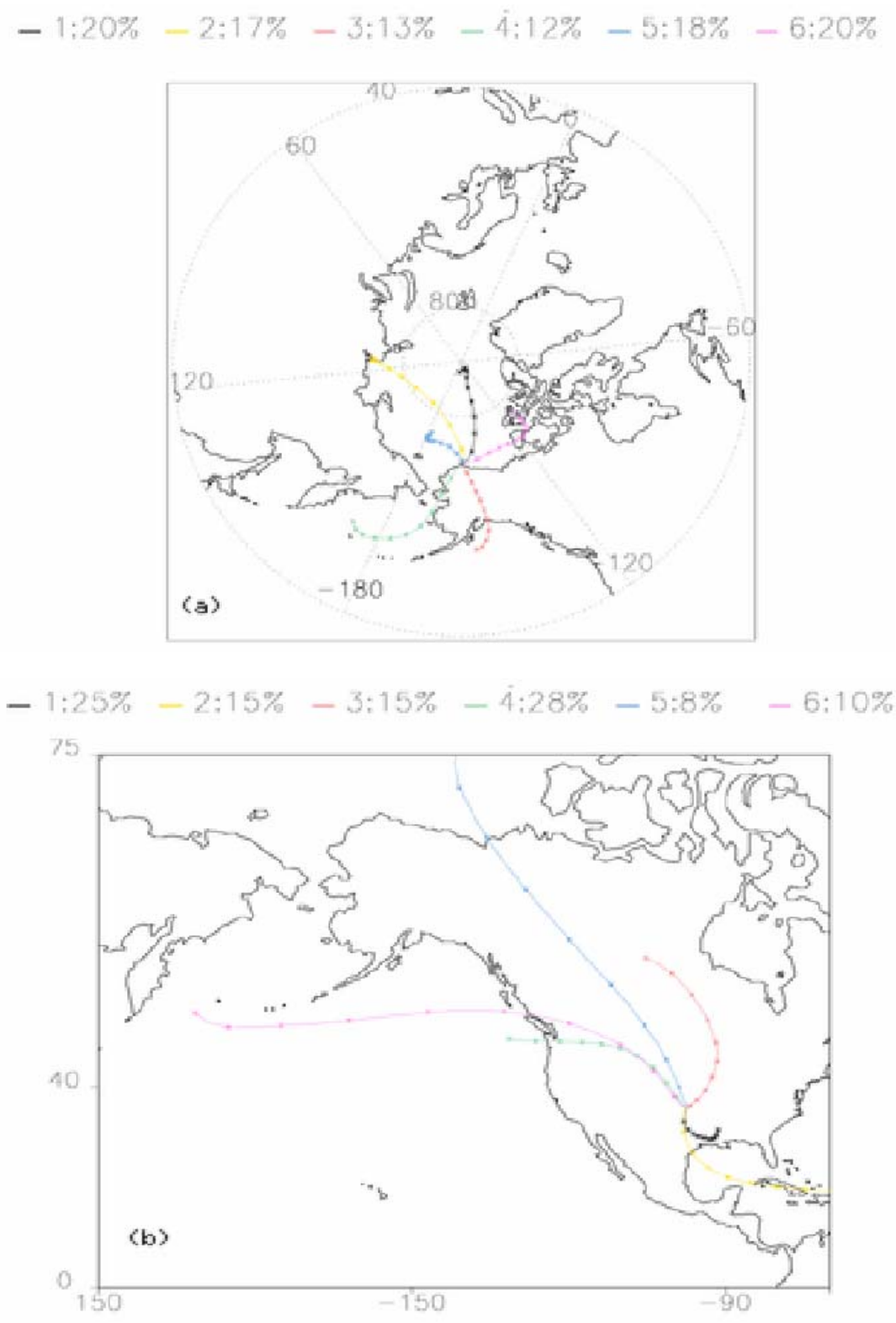

Figure 1. Clusters of 10-day 500 m back trajectories arriving at (a) NSA and (b) SGP.

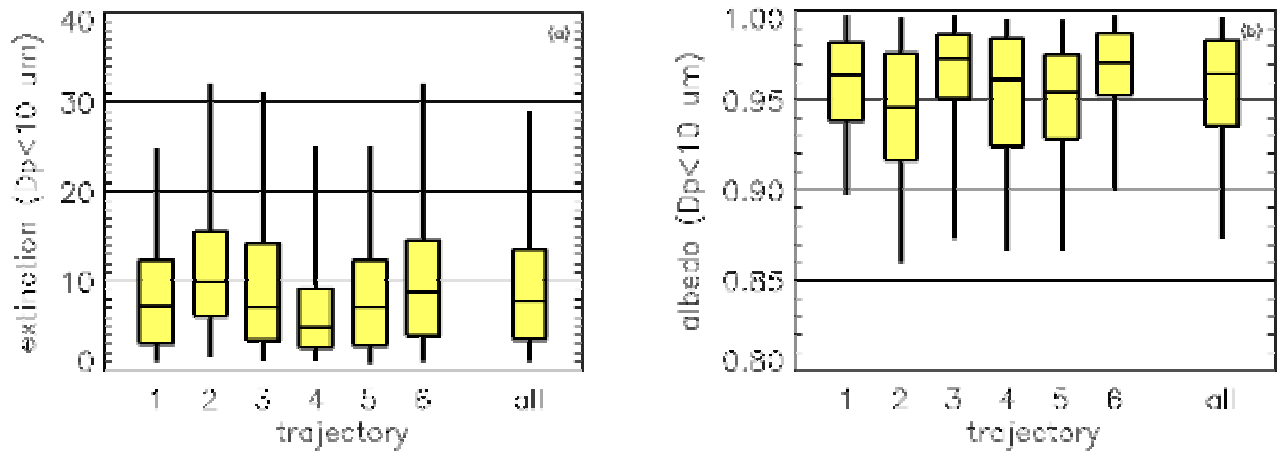

Figure 2. Box whisker plots of (a) aerosol extinction and (b) single scattering albedo as a function of trajectory cluster for NSA.

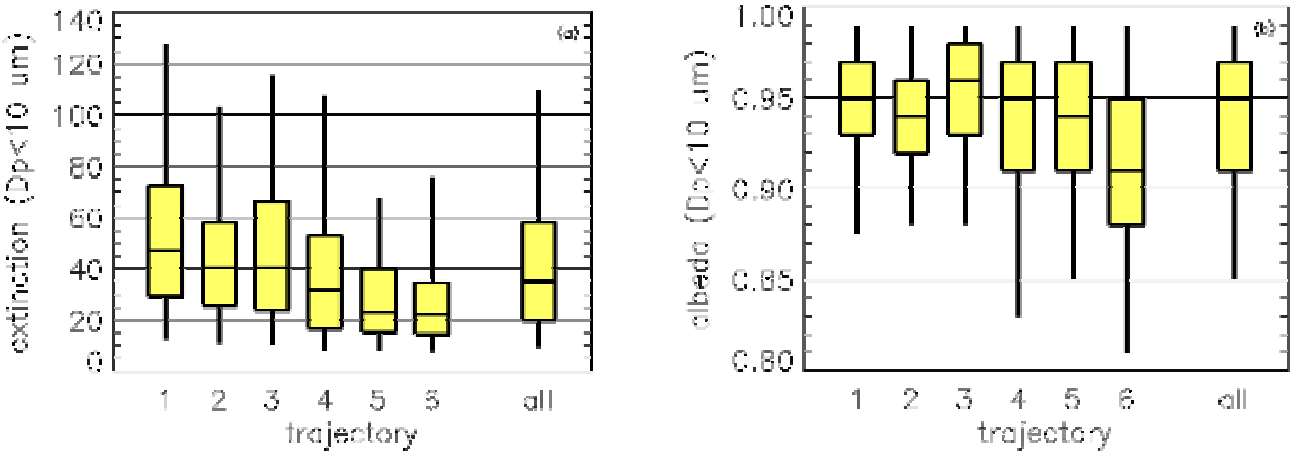

Figure 3. Box whisker plots of (a) aerosol extinction and (b) single-scattering albedo as a function of trajectory cluster for SGP.

The optical and chemical properties observed for each trajectory cluster appear to be consistent with the source region of the trajectory cluster thus illustrating the utility of including airmass trajectory in the statistical evaluation of aerosol types. However, as the statistical plots (Figures 2 and 3) show, there is significant overlap in the values of optical and chemical properties for each trajectory cluster suggesting that airmass trajectory alone may not be adequate for identifying an aerosol type.

Future work, which may help to further constrain the aerosol type, includes the following:

- Incorporate surface humidograph data at the SGP site to provide further information on aerosol chemistry/optics.

- Include local conditions (e.g., wind speed, relative humidity) to help identify periods of local dust contamination or fog, for example, which will influence the optical properties of the sampled aerosol.

- Segment trajectory clusters based on seasonal patterns to remove some of the variability in the observations.

- Utilize in situ aerosol profiles to identify presence of vertical gradients at SGP.

Corresponding Author

John Ogren, John.A.Ogren@noaa.gov

References

Delene, D. J., and J. A. Ogren, 2002: Variability of aerosol optical properties at four North American surface monitoring sites. *J. Atmos. Sci*., **59**, 1135-1150.

Harris, J. M., and J. D. W. Kahl, 1994: Analysis of 10-day isentropic flow patterns for Barrow, Alaska: 1985-1992. *J. Geophys. Res.*, **99**, 25,845-25,855.

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. *J. Geophys. Res.,* **107**, 10.1029/2001JD001248.