

1A1) Investigation of the Optical and Cloud Forming Properties of Pollution, Biomass Burning, and Mineral Dust Aerosol

이 용 섭

기상청 기후예측과

1. Abstract

This thesis describes the use of measured aerosol size distributions and size-resolved hygroscopic growth to examine the physical and chemical properties of several particle classes. The primary objective of this work was to investigate the optical and cloud forming properties of a range of ambient aerosol types measured in a number of different locations. The tool used for most of these analyses is a differential mobility analyzer / tandem differential mobility analyzer (DMA / TDMA) system developed in our research group. To collect the data described in two of the chapters of this thesis, an aircraft-based version of the DMA / TDMA was deployed to Japan and California. The data described in two other chapters were conveniently collected during a period when the aerosol of interest came to us. The unique aspect of this analysis is the use of these data to isolate the size distributions of distinct aerosol types in order to quantify their optical and cloud forming properties.

I used collected data during the Asian Aerosol Characterization Experiment (ACE-Asia) to examine the composition and homogeneity of a complex aerosol generated in the deserts and urban regions of China and other Asian countries. An aircraft-based tandem differential mobility analyzer was used for the first time during this campaign to examine the size-resolved hygroscopic properties of particles having diameters between 40 and 586 nm. Asian Dust Above Monterey (ADAM-2003) study was designed both to evaluate the degree to which models can predict the long-range transport of Asian dust, and to examine the physical and optical properties of that aged dust upon reaching the California coast. Aerosol size distributions and hygroscopic growth are measured in College Station, TX to investigate the cloud nucleating and optical properties of a biomass burning aerosol generated from fires on the Yucatan Peninsula. Measured aerosol size distributions and size-resolved hygroscopicity and volatility were used to infer critical supersaturation distributions of the distinct particle types that were observed during this period. The predicted CCN concentrations were used in a cloud model to determine the impact of the different aerosol types on the expected cloud droplet concentration. RH-dependent aerosol extinction coefficients are calculated at a wavelength of 550 nm.

2. Summary and conclusions

In this thesis, I describe a method for examining the optical and cloud forming properties of specific classes of aerosols. The underlying objective of this work is to contribute to an improved understanding of the direct and indirect effects of aerosols on climate. In Chapter 2, I describe use of data collected during the Asian Aerosol Characterization Experiment (ACE-Asia) to examine the composition and homogeneity of a complex aerosol generated in the deserts and urban regions of China and other Asian countries. An aircraft-based tandem differential mobility analyzer was used for the first time during this campaign to examine the size-resolved hygroscopic properties of particles having diameters between 40 and 586 nm. Hygroscopic growth factors at 90% RH typically exceeded 1.4, suggesting the soluble fraction of the aerosol was relatively high. The mean standard

deviation of the measured growth factor distributions, which is related to the compositional heterogeneity of the aerosol, increased with increasing particle size, although this trend often reversed for the largest particles considered. Overall, hygroscopicity was observed to increase with proximity to the surface, and with increasing size. One cause of the observed height dependence is the frequent presence of non-hygroscopic dust particles at higher levels that, closer to the surface, were either not observed or had been coated with soluble compounds such as sulfate.

Chapter 3 describes results from the Asian Dust Above Monterey (ADAM-2003) study designed both to evaluate the degree to which models can predict the long-range transport of Asian dust, and to examine the physical and optical properties of that aged dust upon reaching the California coast. By coupling the measured size distributions with a size-dependent fractional categorization based on the hygroscopic growth measurements, independent size distributions for the dust and other aerosol types were created. Within the sampled layers, both the overall mass concentration and light extinction coefficient were dominated by the larger dust particles. Light scattering coefficients predicted based on the measured size distributions compared well with those directly measured with a nephelometer. The calculated mass extinction efficiency of the dust aerosol was somewhat higher than that observed during other studies suggesting that the size distribution had shifted towards smaller size as gravitational settling removed the largest dust particles.

Chapter 4 describes use of aerosol size distributions and hygroscopic growth measured in College Station, Texas to investigate the cloud nucleating properties of a biomass burning aerosol generated from fires on the Yucatan Peninsula. Whereas most other studies designed to investigate biomass burning aerosol have been conducted close to the aerosol source, the data described in this chapter provide details of the properties of an aged smoke aerosol. Measured aerosol size distributions and size-resolved hygroscopicity and volatility were used to infer critical supersaturation distributions of the distinct particle types that were observed during this period. The average inferred CCN concentrations were 869, 1918, and 2206 (cm^{-3}) at 0.15, 0.5, and 1.0% critical supersaturations, respectively. The predicted CCN concentrations were used in a cloud model to determine the impact of the different aerosol types on the expected cloud droplet concentration. For certain aerosol measurements and cloud updraft velocities, predicted cloud droplet concentrations were lower when the observed sulfate aerosol was considered together with the biomass burning aerosol relative to that when only the biomass burning aerosol was considered. By suppressing activation of the less hygroscopic biomass burning particles, pollution aerosols may actually help preserve the smoke aerosol by reducing wet deposition rates. Furthermore, this represents a scenario in which pollution aerosol may cause a slight reduction in cloud albedo.

Chapter 5 describes use of the measurements discussed in Chapter 4 to examine the optical properties of the smoke-dominated aerosol. As was done to isolate the CCN spectra of the biomass burning aerosol, the collected hygroscopicity data were used to separate the sparingly hygroscopic smoke particles from other aerosol types. The approach used to isolate the size distributions of each of the particle types identified permits examination of the optical properties of biomass burning particles even in conditions in which other particle types are more abundant. The size distributions of the smoke and other particle types, coupled with the aerosol properties derived from the TDMA data, were used to calculate RH-dependent aerosol extinction coefficients at a wavelength of 550 nm. This provides a method for characterizing changes in biomass burning aerosols that occur over periods of days, which would typically not be possible due to the accumulation of additional particle types over time.