MIXING STATE, GROWTH KINETICS AND AGING OF AMBIENT POLLUTED CCN

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INTRODUCTION

We present an overview of Cloud Condensation Nuclei (CCN) measurements in the vicinity of Houston, TX during the GoMACCS/TEXAQS campaign (August-September, 2006), and in Mexico City during the MILAGRO campaign (March, 2006). Measurements in Houston were obtained aboard the NOAA P3 and the CIRPAS Twin Otter platforms, and polluted air masses in and out of cloudy regions were sampled over a total of 40 flights. Measurements in Mexico City were ground-based, using a DMT Cloud Condensation Nucleus counter (CCNc) that sampled classified aerosol from a Differential Mobility Analyzer (DMA) to obtain the size-resolved activation fraction as a function of water vapor supersaturation (SS).

The wide range of CCN concentration, aerosol composition and aging/mixing state makes these particularly valuable datasets for constraining uncertainties associated with prediction of CCN concentration and cloud droplet number for polluted clouds.

ASPECTS OF THE HOUSTON DATASET

Ambient aerosol in Houston, Texas was sampledonboard the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter during the Gulf of Mexico Atmospheric Composition and Climate Study (GoMACCS) field campaign (August 25-September 15, 2006). Aersol was also sampled onboard the NOAA WP-P3 during the Texas Air Quality Study (TexAQS). CCN sampled originated from a variety of local sources as petrochemical industries, refineries, power plants, ship yards, and the urban Houston plume. Figure 1 shows the flight tracks for the Twin Otter Research Flights (RF), all of which occurred during the daylight hours.

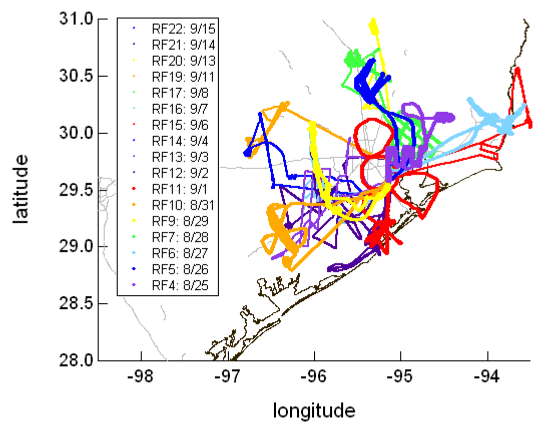

Figure 1. Research flight tacks conducted with the CIRPAS Twin Otter.

The concentration of CCN was measured using a Droplet Measurement Technologies continuous flow streamwise thermal gradient CCN counter (CCNc), operated between 0.1 and 0.4% supersaturation. CCN closure was obtained using the particle size distribution measured by the Dual Automated Classified Aerosol Detector (DACAD) and the aerosol chemical composition measured by an Aerodyne Aerosol Mass Spectrometer (AMS). CCN concentrations ranged from 100 cm^{-3} to more than 10,000 cm^{-3}. The aerosol was typically composed of a mixture of organic and sulfate; organic mass fraction in the aerosol ranged from 10 to 90%.

Figure 2 shows the predicted vs. measured CCN concentrations, assuming that particles are composed purely of ammonium sulfate; data is presented for all the research flights in Figure 1. On average, the CCN closure is remarkably good, given the very simple assumption on particle chemistry; CCN concentrations were on average overpredicted by 20%. Explicit consideration of chemical composition further improves closure (not shown). The same aspects in the CCN closure were also seen in the P3 dataset.

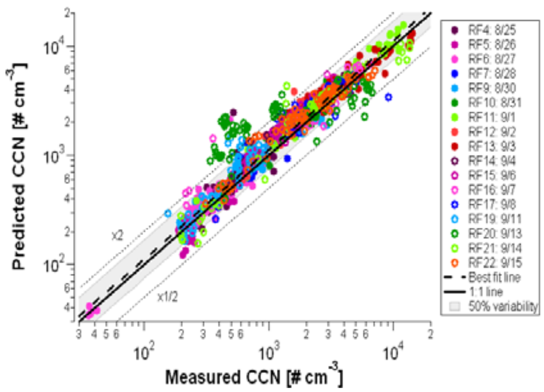

Figure 2. CCN Closure for all research flights conducted with the CIRPAS Twin Otter, where particles are assumed to be composed of ammonium sulfate.

ASPECTS OF MEXICO CITY DATASET

MIRAGE, a part of a larger international study MILAGRO (Megacity Initiative: Local and Global Research Observations), encompassed measurements from several aircraft and ground-based sites. Our measurements focused on the CCN activity of ambient aerosol downwind of Mexico City at the University of Tecámac ground site. The measurements were obtained at the peak of the dry season, from March 16 - April 1, 2006. The aerosol sampled were impacted by strong dust events, heavy rains, local biomass burning, vehicular emissions from a nearby roadway, extremely high ammonium concentrations and the Mexico City plume. On occasion, emissions from the Tula power plant (~40 km away) impacted the aerosol sampled.

Size-resolved CCN activity measurements were used to determine the "activated fraction," or the fraction of particles that activate into cloud droplets as a function of water vapor supersaturation and dry particle size. These measurements, combined with Köhler theory, are used to determine the fraction of particles that act as CCN, and, infer the distribution of soluble volume fraction (SVF) for the CCN-active fraction as a function of time and dry particle size (Lance, 2008). Contours of SVF for 100 nm particles are shown in Figure 3.

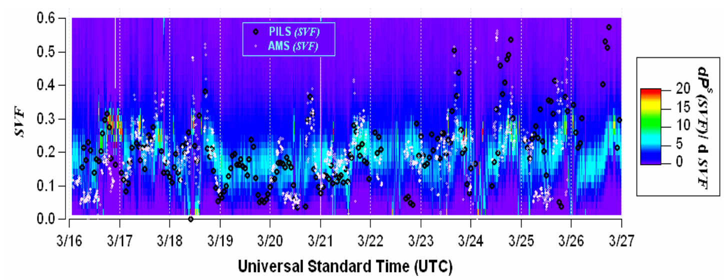

Figure 3. Contours of SVF for the data of this study. Superimposed are SVF obtained from PILS and an aerosol mass spectrometer.

Inferred SVF agrees well with direct measurements of composition (PILS, AMS), (except when the amount of dust is high) and affirms the applicability of the method.

Inferred SVF is relatively insensitive to wind direction, rain, dust, or local biomass burning events; suggesting that the aerosol retain the characteristics of the larger-scale "background" particulate matter. The most obvious trend in SVF is a diurnal pattern, where SVF tends to be maximum (and with a narrow distribution) during the daytime, likely due to condensation of hydrophilic compounds (photochemically generated) onto aerosol particles.

REFERENCES

Lance, S, PhD Thesis, Georgia Institute of Technology, 2008.

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