Appendix B:
Toward Aerosol/Cloud Condensation Nuclei (CCN) Closure During CRYSTAL-FACE*

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 108, NO. D20, 4633, doi:10.1029/2003JD003582, 2003

Toward aerosol/cloud condensation nuclei (CCN) closure during CRYSTAL-FACE

Timothy M. VanReken,[1] Tracey A. Rissman,[1] Gregory C. Roberts,[2]
Varuntida Varutbangkul,[1] Haflidi H. Jonsson,[3] Richard C. Flagan,[1] and John H. Seinfeld[1]

Received 10 March 2003; revised 1 July 2003; accepted 15 July 2003; published 18 October 2003.

[1] During July 2002, measurements of cloud condensation nuclei were made in the vicinity of southwest Florida as part of the Cirrus Regional Study of Tropical Anvils and Cirrus Layers-Florida Area Cirrus Experiment (CRYSTAL-FACE) field campaign. These observations, at supersaturations of 0.2 and 0.85%, are presented here. The performance of each of the two CCN counters was validated through laboratory calibration and an in situ intercomparison. The measurements indicate that the aerosol sampled during the campaign was predominantly marine in character: the median concentrations were 233 cm^{-3} (at $S = 0.2\%$) and 371 cm^{-3} (at $S = 0.85\%$). Three flights during the experiment differed from this general trend; the aerosol sampled during the two flights on 18 July was more continental in character, and the observations on 28 July indicate high spatial variability and periods of very high aerosol concentrations. This study also includes a simplified aerosol/CCN closure analysis. Aerosol size distributions were measured simultaneously with the CCN observations, and these data are used to predict a CCN concentration using Köhler theory. For the purpose of this analysis, an idealized composition of pure ammonium sulfate was assumed. The analysis indicates that in this case, there was good general agreement between the predicted and observed CCN concentrations: at $S = 0.2\%$, $N_{predicted}/N_{observed} = 1.047$ ($R^2 = 0.911$); at $S = 0.85\%$, $N_{predicted}/N_{observed} = 1.201$ ($R^2 = 0.835$). The impacts of the compositional assumption and of including in-cloud data in the analysis are addressed. The effect of removing the data from the 28 July flight is also examined; doing so improves the result of the closure analysis at $S = 0.85\%$. When omitting that atypical flight, $N_{predicted}/N_{observed} = 1.085$ ($R^2 = 0.770$) at $S = 0.85\%$. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0320 Atmospheric Composition and Structure: Cloud physics and chemistry; 1610 Global Change: Atmosphere (0315, 0325); *KEYWORDS:* CCN closure, CCN instrumentation, CRYSTAL-FACE, aircraft measurements, cloud

Citation: VanReken, T. M., T. A. Rissman, G. C. Roberts, V. Varutbangkul, H. H. Jonsson, R. C. Flagan, and J. H. Seinfeld, Toward aerosol/cloud condensation nuclei (CCN) closure during CRYSTAL-FACE, *J. Geophys. Res.*, *108*(D20), 4633, doi:10.1029/2003JD003582, 2003.

1. Introduction

[2] The importance of clouds in the climate system is well established; clouds play a vital role in the global radiation budget and hydrological cycle. Clouds form when a parcel of air becomes supersaturated with respect to water vapor and the excess water condenses rapidly on ambient particles to form droplets. For this rapid condensation (termed activation) to occur at a given supersaturation, the particle must have sufficient soluble mass; this subset of the aerosol population is called cloud condensation nuclei, denoted CCN. The atmospheric concentration of CCN is often substantially enhanced by human activities, and the various ways that this enhancement affects the radiative properties of clouds are collectively known as indirect aerosol forcing (the inclusion of the word ''indirect'' differentiates these effects from the direct aerosol effect, which describes the radiative interactions of the particles themselves). Cloud processes are complex by nature and heavily dependent on purely dynamical factors, but in general terms indirect aerosol effects can be split into two categories. For a given supersaturation, an air mass with a higher CCN concentration would produce a cloud with a higher droplet concentration, but a smaller mean droplet diameter; this often results in a more reflective cloud and is known as the first indirect effect or Twomey effect [*Twomey*, 1977]. The second indirect effect, identified by *Albrecht* [1989], also stems from the smaller average droplet diameter in polluted clouds; a smaller mean droplet size inhibits the processes

[1]Department of Chemical Engineering, California Institute of Technology, Pasadena, California, USA.
[2]Center for Atmospheric Sciences, Scripps Institution of Oceanography, San Diego, California, USA.
[3]Center for Interdisciplinary Remotely Piloted Aircraft Studies, United States Naval Postgraduate School, Marina, California, USA.

0148-0227/03/2003JD003582$09.00

that lead to precipitation, thereby increasing cloud lifetime and therefore cloud coverage.

[3] While observations support the existence of indirect aerosol effects on a local scale [*Johnson et al.*, 1996; *Rosenfeld*, 1999, 2000; *Durkee et al.*, 2000; *Garrett et al.*, 2002], current understanding of the processes involved is insufficient to accurately predict the global importance of indirect aerosol forcing. The *Intergovernmental Panel on Climate Change* (*IPCC*) [2001] estimates that the first indirect effect results in a global mean forcing of between 0 and -2 W m^{-2} and does not give an estimate for the second indirect effect, which is also expected to be one of cooling. Reliable predictions regarding climate forcing await more detailed understanding of the dependence of cloud properties on aerosol properties.

[4] The first step in understanding the relationship between the ambient aerosol and the cloud that forms therefrom is to know the activation properties of the atmospheric aerosol. In theory, if a particle's size and chemical composition were precisely known, the supersaturation at which activation occurs could be calculated using Köhler theory [*Seinfeld and Pandis*, 1998]. However, ambient aerosol populations can contain myriad chemical species, the activation properties of most of which have not been established. Furthermore, recent studies have demonstrated that simply categorizing aerosol species into soluble and insoluble fractions is sometimes insufficient [*Cruz and Pandis*, 1998; *Hegg et al.*, 2001; *Raymond and Pandis*, 2002]; slightly soluble species, surfactants, and soluble gases can affect activation either thermodynamically or kinetically [*Charlson et al.*, 2001; *Nenes et al.*, 2002]. To establish the connection between theory and the actual atmosphere, it is desirable to directly measure the portion of the aerosol population that activates at a given supersaturation. Such a measurement generally involves exposing an aerosol sample to a known supersaturation; the CCN active at that supersaturation rapidly grow to a size at which they can be counted by standard techniques. In the laboratory, instruments using such measurement strategies can be tested using aerosols whose size and chemical properties are carefully controlled. Then, the activation behavior of an ambient aerosol can be measured, giving rise to a so-called closure experiment, whereby measured CCN concentrations are compared against predictions based on simultaneously measured aerosol size and composition data. A successful closure study serves to validate both the performance of the CCN instrument itself and the theoretical basis for the prediction of the activation properties of the aerosol.

[5] The Cirrus Regional Study of Tropical Anvils and Cirrus Layers-Florida Area Cirrus Experiment (CRYSTAL-FACE) field campaign in the Florida Keys during July 2002 had as its goal the investigation of the properties of tropical convective systems and the resultant cirrus layers. These cirrus layers, known as anvils, affect the radiative balance [*Ramanathan et al.*, 1989], and a detailed understanding of the physical processes involved in their formation would enhance the ability to predict their occurrence and lifetime. As part of the CRYSTAL-FACE campaign, the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter aircraft flew 20 research missions, focused on characterizing the ambient aerosol in the vicinity of the convective systems, measuring cloud properties, and on making radiation measurements below the cirrus anvils. Data were collected both over land and water along the southwest coast of Florida; Figure 1 shows the flight tracks for the missions for which CCN data are available. Table 1 provides details on each research flight, and Table 2 lists the aerosol and gas-phase instrumentation on board the Twin Otter.

[6] This study presents the airborne CCN measurements from CRYSTAL-FACE and examines the extent to which it is possible to predict CCN concentrations from size distribution data in the absence of a detailed knowledge of the aerosol composition. Two CCN counters were on board the Twin Otter (Table 2). One instrument, operating at a supersaturation of approximately 0.85%, provided useful data for all but three flights, when electrical noise from another instrument caused the CCN counter to malfunction. The second CCN counter, with an effective supersaturation of approximately 0.2%, was operated for all but one flight from CF-8 through the end of the campaign; no data are available from CF-16 due to an instrument malfunction. The reliability of these measurements is verified by laboratory experiments, by a field intercomparison of the two instruments, and by comparison with other instruments measuring aerosol concentration. After establishing the validity of the data, the observations are described in more detail in order to provide a comprehensive picture of the typical summertime CCN population over southwest Florida. A simplified closure analysis follows, comparing the CCN data set at both measured supersaturations to size spectral data from the Caltech differential mobility analyzer (DMA, described by *Wang et al.* [2003]), assuming an idealized (ammonium sulfate) composition. The study concludes by discussing the sensitivity of the results to assumptions made in the analysis.

2. Background

[7] Previous attempts to match predicted CCN concentrations with those directly observed have met with mixed success. The methods by which these studies were conducted vary considerably, and by examining the details of these methodologies one can determine those elements required for a successful experiment.

[8] Only three studies in the literature present results that can be considered successful in terms of aerosol/CCN closure. All were ground-based studies: *Liu et al.* [1996] made measurements in Nova Scotia as part of the North Atlantic Regional Experiment (NARE), *Cantrell et al.* [2001] used measurements made in the Maldives during the Indian Ocean Experiment (INDOEX), and *Roberts et al.* [2002] collected data in the Amazon Basin during the Cooperative Large-Scale Biosphere-Atmosphere (LBA) Airborne Regional Experiment 1998 (CLAIRE-98). In the first two studies, the aerosol was split into soluble and insoluble fractions based on filter samples and the soluble fraction was assumed to be ammonium sulfate. Roberts et al. further split the soluble fraction into organic and inorganic components. All three studies averaged the CCN and size spectral data over a substantial period of time to match the filter sampling time. Liu et al. used an isothermal haze chamber to obtain CCN concentrations at a supersaturation,