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The Interaction of Water and Aerosols in the Marine Boundary Layer: A Study of Selected Processes Impacting Radiative Transfer and Cloudiness

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Awards #: N00014-07-1-0277
http:// http://www.atmos.washington.edu/academic/atmoschem.html

LONG-TERM GOALS

The overarching, long-term goal of the study is to explore the profound effect of aerosol-water interaction both on radiation propagation in, and the thermodynamic structure of, the marine boundary layer. Specific goals are: 1) compile a climatology of aerosol hygroscopicity for use in the NAAPS and COAMPS models, and, further, to develop a model parameterization of hygroscopicity based on aerosol size and composition for such models, 2) explore the relative impacts of cross-inversion mixing and sub-cloud aerosol on cloud thickness and cloud base height, 3) quantify and parameterize the impact of precipitation scavenging on below cloud radiative transfer and cloud liquid water path. The sampling platform utilized is the CIRPAS Twin Otter research aircraft and the venue is the littoral environment off the California coast, representative of areas with high shipping densities.

OBJECTIVES

For the current reporting period, our efforts have centered on completing several different analyses utilizing data both from the various CARMA studies, the VOCLAS study and additional studies such as RED, ACE-Asia and SAFARI, the precise suite of data bases being dependent on the specific analysis. Our objectives for these analyses have changed somewhat from those in our original proposal in light of our findings to date. We summarize them as follows.

- Determine the relationship between cloud drop number concentration (CDNC) and the properties of the precursor aerosol that have the most prognostic power using data from the most important cloud venues from the standpoint of cloud radiative climate forcing.

- Perform a feasibility analysis of using arctic deposition data to test the NAAPS model.
- Assess the importance of elemental carbon (EC) particles as CCN using data from both the CARMA studies and, if feasible, VOCALS.
- Develop a climatology of aerosol optical properties based on data from the CARMA field studies plus data from various other field campaigns.

APPROACH

The first objective involves data on CDNC and the various properties of precursor aerosols most indicative of cloud drop activation. The obvious proxy would be CCN concentrations and these are in fact measured with one of two instruments. The first and most widely used is the DMT CCN-100 spectrometer while the second is the University of Wyoming's MA-100 static diffusion chamber. However, CCN actually have little prognostic power without accompanying measurements of cloud supersaturation, a parameter that cannot be directly measured. It has been proposed that essentially aerosol size alone can more simply be used to predict CDNC (cf., Dusek et al, 2006). While this has been challenged (e.g., Hudson, 2007) and is likely not universally true, it may well be true for the three main stratocumulus decks of the world (off the coasts of California, Chile and Namibia, respectively), that largely control aerosol indirect forcing. We assume as a working hypothesis that the accumulation mode number concentration (AMNC) is a viable proxy for effective CCN, essentially CDNC, for our venues. This parameter is measured with a PMS/DMT PCASP-100x. Comparisons between the CDNC and AMNC are explored with regression analysis.

The second objective is addressed by a comparison of measured deposition of both precipitation and various chemicals in that precipitation derived from several recent studies of deposition of light absorbing aerosols in arctic snow (Hegg et al, 2009; 2010), with model estimates of such deposition for the sampling locations and times of deposition.

The methodology for achieving the third objective involves the use of two instrument new to our program in addition to the PCASP mentioned above. The first of these is the annular geometry CCN spectrometer manufactured by DMT Inc. to which we also alluded above. This will yield a continuous record of the CCN concentration at five supersaturations with a time resolution of about 10 minutes. These data, and concurrent measurements of the AMNC, will be compared with the concentration of EC bearing particles measured by the SP2 instrument manufactured by DMT Inc. and recently evaluated and described by Moteki and Kondo (2007). The comparison will yield the fraction of the AMNC that contain EC and possibly, if a sufficient number of long enough horizontal flight legs can be found, CCN number concentration at each supersaturation that contain EC.

The final objective, the development of a climatology of aerosol optical properties is addressed in a straightforward manner using standard statistical techniques to derive the distributions of the various optical properties over the various field campaigns for which we have data.

WORK COMPLETED

To date, work on the first objective is well along and a manuscript is currently being prepared for submission. Work on the second objective has been completed and the results relayed to our colleagues at NRL Monterey (who operate the NAAPS model) for their consideration. Work on the third objective

has proceeded slowly, as discussed in previous reports but some analysis and results are now in hand. Work on the fourth objective was temporarily stalled but has now resumed.

RESULTS

For the regression analysis of the CDNC-aerosol relationship, data were derived from two years of CARMA data (2005, 2007), and from the VOCALS-Rex study, associated with the stratocumulus decks of California and Chile, respectively. While no data gathered directly by us were available for the third of the main global stratocumulus decks, that off of the coast of Namibia, a limited amount of data was gleaned from either the literature (e.g., Kiel and Haywood, 2003) or from public data archives (the archive for SAFARI 2000 data in the CARG archive at the University of Washington). For a direct comparison of CDNC with CCN active at various supersaturations (0.2 to 1.0 %), no relationship with an R^2 in excess of 0.33 could be found. However, for the CDNC-AMNC relationship, an excellent regression fit was found and is shown in Figure 1. The R^2 of 0.9 exceeds anything of which we are aware from previous work. It suggests that AMNC is a very powerful prognostic parameter for the venues examined and could provide a very useful tool both in remote retrieval of effective CCN and for use in large scale climate models.

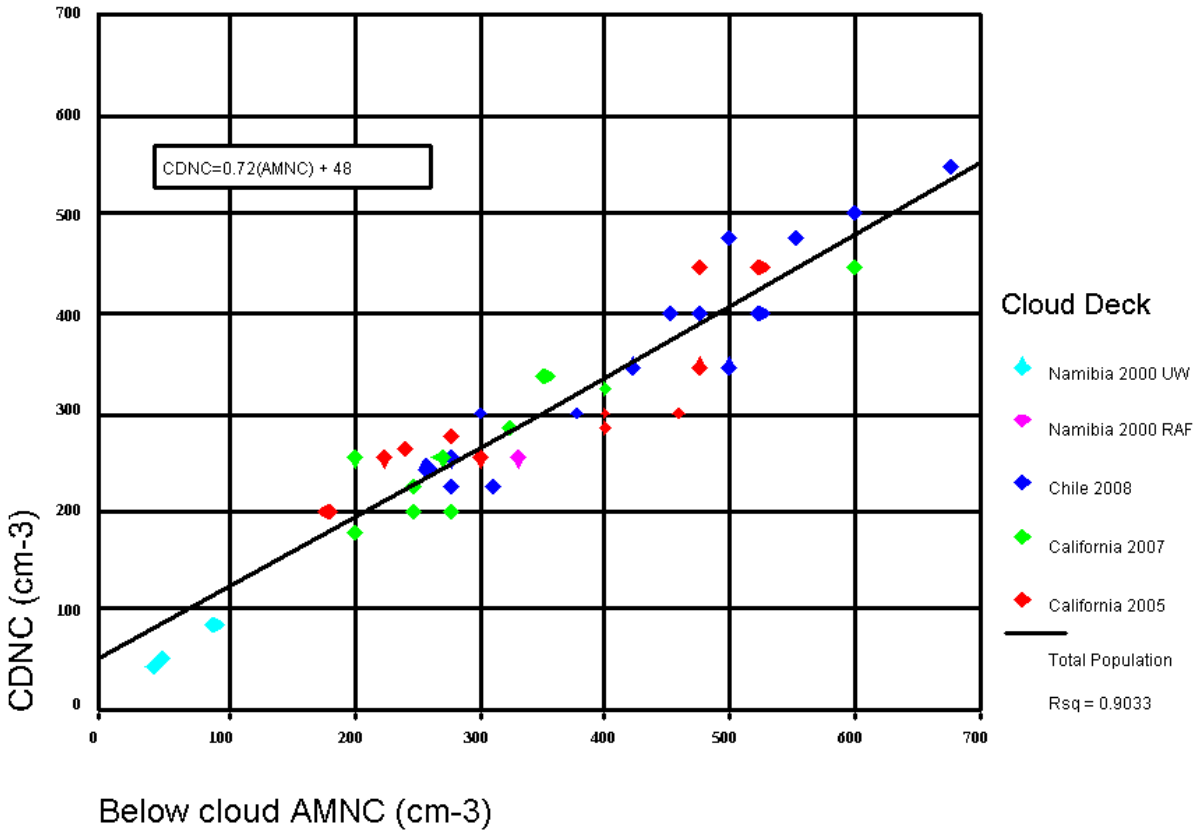

Figure 1 Regression analysis of the dependence of the CDNC on the AMNC for the three main stratocumulus decks of the earth system. Note that RAF refers to the data from Kiel and Haywood (2003) while UW refers to data from the University of Washington CARG archive.

Comparison of various NAAPS variables associated with deposition to observations of the same quantities derived from several studies of aerosol deposition in the arctic (alluded to above) has suggested some systematic problems with the NAAPS model. For example, the fundamental comparison of precipitation amount, illustrated in Figure 2, suggests that the model is systematically overestimating precipitation in the arctic venue. On the other hand, a similar analysis for NSS sulfate (not shown) suggests a large underprediction of the deposition of this species, in accord with internal assessments at NRL Monterey. To explore this further, we participated in a study to assess the role of various in-cloud sulfate production mechanisms in the marine sulfur budget. We will use the information from this study to assess whether or not NAAPS includes proper sulfate chemistry. While not definitive, these analyses do demonstrate the feasibility of using available observational data in assessment of the NAAPS model.

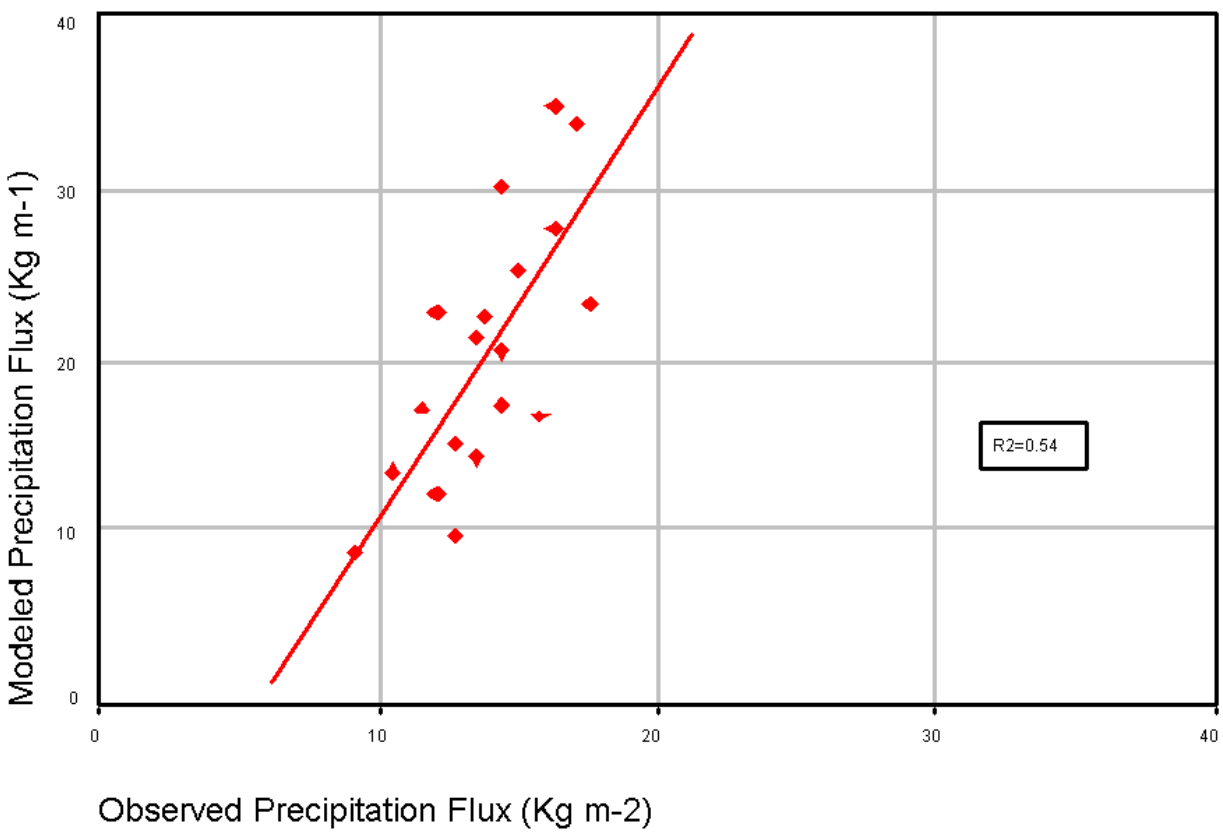

Figure 2. Regression of precipitation fluxes predicted by the NAAPS model with observations of the flux derived from snow depth measurements in the arctic. The correlation between observed and predicted fluxes is modest and the model appears to over predict the flux by ~ 60%.

The assessment of the importance of EC-containing particles as CCN has proven troublesome. The primary reason for this is the limited database, the available measurements being confined to the CARMA IV study (2007) and VOCALS (2008). Within this data base, there is substantial variability in the fraction of potentially cloud active particles that contain EC. This can be seen in Figure 3, in which

typical co-measured concentrations of AMNC and EC-containing particles are plotted for both the CARMA and VOCALS studies (note that, as per the analysis summarized in Figure 1, AMNC is the parameter of choice for effective CCN concentration). For CARMA, while occasionally the EC particle concentration is comparable to the AMNC, it is usually less than 10% of the AMNC, suggesting no particularly significant role for the EC particles, certainly not a true modulating influence. For the more polluted VOCALS study area, on the other hand, the EC particles are commonly a more substantial fraction of the AMNC, on the order of 50% of the AMNC concentration. For the third stratocumulus deck of global significance, that off Namibia, the study by Kiel and Haywood (2003) suggests that biomass burning plumes will commonly advect over the offshore stratocumulus deck and these plumes do contain a relatively high number of EC particles – as do such plumes in the two stratocumulus regions for which we have data. However, the extent of the interaction of the fire plumes with the essentially boundary layer stratocumulus is not clear from the data published to date. Similarly, for the three regions assessed here taken as a whole, the extent to which EC particles modulate CDNC is not clear. Certainly there are instances when this is likely the case but clearly at least as many more in which it is not. A much more extensive database will be necessary for a definitive assessment.

The last objective of this study, the aerosol optical climatology, is now well along, roughly 75% completed. We should finish it in November of this year.

IMPACT/APPLICATIONS

The CDNC-AMNC relationship shown in Figure 1 should have a substantial impact on the ability of remote sensing to characterize effective CCN concentrations for the important stratocumulus deck regions of the atmosphere. It should also be valuable for use in large-scale models that deal with indirect aerosol forcing. The establishment of the feasibility of using available deposition data to assess NAAPS model performance should, of course, aid in the refinement of the NAAPS model, as should the completion of the aerosol optical climatology.

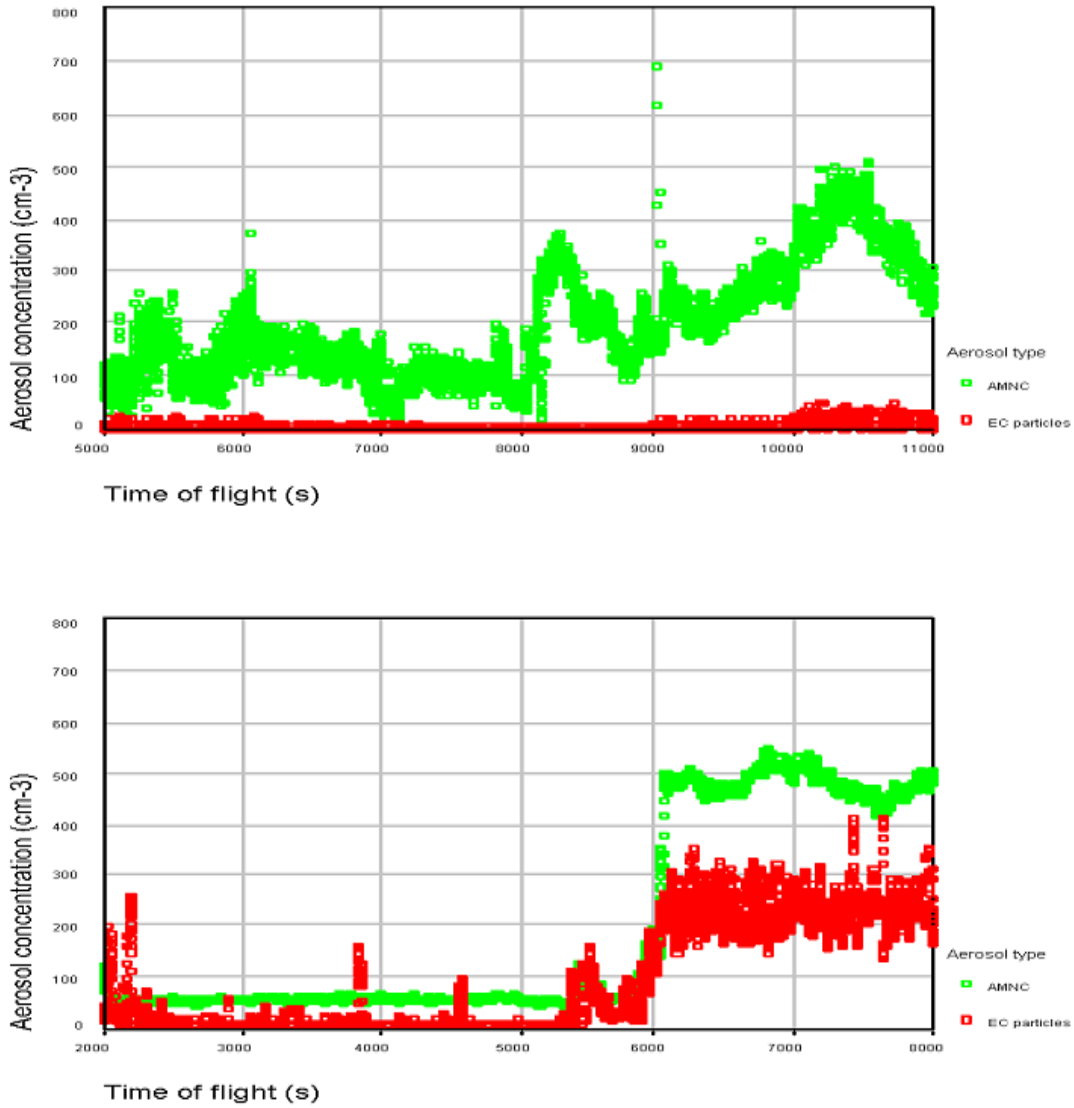

Figure 3. Examples of the portion of AMNC that contain EC for both the CARMA (a) and VOCALS (b) studies. Note that the abrupt increase in concentrations shown in (b) corresponds to a transition of the sampling aircraft from the free troposphere down into the marine boundary layer.

TRANSITIONS

None.

RELATED PROJECTS

These measurements are highly relevant to determination of aerosol light scattering in the MBL (and thus radiative transfer in the MBL), and CCN activity (and thus of the microphysics of MBL clouds). Furthermore, numerical transport models could now incorporate a simple relationship such as that

shown in Figure 1, even for a limited domain, and thus improve their prognostic power for cloud optical properties.

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