Measurements of the Aerosol Light-Scattering Coefficient at Ambient and 85% Relative Humidity on the ONR Pelican During ACE-2

Dean A. Hegg
University of Washington
Department of Atmospheric Sciences
Box 351640
Seattle, WA 98195-1640
phone: (206) 685-1984 fax: (206) 685-7160 email: deanhegg@atmos.washington.edu

David S. Covert
University of Washington
Department of Atmospheric Sciences
Box 354235
Seattle, WA 98195-4235
phone: (206) 685-7461 fax: (206) 685-3397 email: dcovert@u.washington.edu

Award Number: N00014-97-1-0132
http://cargsun2.atmos.washington.edu/~santiago/ACE2/html/2panel_page.html

LONG-TERM GOALS

The ultimate goal of this study, as originally conceived, is the determination of aerosol hygroscopicity and its impact on physical (e.g., size) and optical (e.g., scattering coefficient) properties of marine aerosols on large spatial scales. In the course of our analysis, it has further become clear that relating aerosol hygroscopicity to chemical composition, particularly organic composition, is an integral component of this research. Additionally, associated with the renewal of funding in 2002, goals concerning the interaction of marine aerosols with marine boundary layer (MBL) clouds have been added. These new goals include: 1) assessment of the relative impacts of purely meteorological and aerosol factors on cloud albedo, 2) determination of the impact of very large CCN on cloud albedo, 3) assessment of the impact of cloud processing on aerosol light scattering, and 4) explore, observationally, the impact of organics on CCN activation in MBL clouds.

OBJECTIVES

Several of the objectives enumerated in previous reports have now been largely achieved (e.g., assessment of the impact of humidity on aerosol scattering coefficient for different aerosol types in ACE-2, remote retrieval of aerosol hygroscopicity and CCN concentration). During the current year, our efforts have centered on analysis of the data gather during the RED and CARMA-I field campaigns. Our objectives for these campaigns focus on the following.

- Explore the relationship between aerosol hygroscopicity and organic composition using the Ming/Russell thermodynamic model (POEM), coupled with Mie scattering model calculations, to relate the hygroscopicity as a function of size generated by POEM with observed aerosol light scattering.

Report Documentation Page

Form Approved
OMB No. 0704-0188

1. REPORT DATE	2. REPORT TYPE	3. DATES COVERED
30 SEP 2003		**00-00-2003 to 00-00-2003**

4. TITLE AND SUBTITLE	5a. CONTRACT NUMBER
Measurements of the Aerosol Light-Scattering Coefficient at Ambient and 85% Relative Humidity on the ONR Pelican During ACE-2	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **University of Washington,,Department of Atmospheric Sciences,Box 351640,,Seattle,,WA,98195**	8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES

14. ABSTRACT

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT **unclassified**	b. ABSTRACT **unclassified**	c. THIS PAGE **unclassified**	**Same as Report (SAR)**	**7**	

- Explore the impact of cloud processing on aerosol radiative properties, including the aerosol mass scattering efficiency.

- Explore the impact of aerosols on cloud albedo and compare this with the impact of such purely meteorological parameters as the Sea Surface Temperature (SST).

APPROACH

The technique used to quantify the hygroscopicity of aerosols from in-situ measurements has been described in previous reports. Concurrent analyses of the chemical composition of the aerosol, and in particular the organic composition, are now routinely done. Aerosol samples are first obtained by either filtration or impaction (filter packs or MOUDI impactors) and subjected to an array of analytical techniques upon return to the laboratory. These include ion chromatography with both conductivity and pulsed amperometric detectors, and LC-MS. Relating the analyzed aerosol composition to the hygroscopicity has been done in the past on an empirical basis via multiple regression and factor analysis. In the current period, we have turned additionally to a more fundamental approach utilizing a variant of the Ming/Russell aerosol model (Ming and Russell, 2001), now labeled POEM. Output from this model, which predicts the equilibrium size of aerosol particles at a given RH based on their chemical composition is used as input for Mie scattering code which yields the light-scattering coefficient for the given aerosol composition and size distribution. Such predications can be compared with our humidigraph measurements.

Methodologies for analysis of the data relevant to the second objective – the impact of cloud processing on aerosol radiative properties – are not so much instrumental as procedural, i.e., which variables to choose and how to compare them. For example, one must determine what air has been recently cloud processed and what has not. We do this by means of an analysis of the thermodynamic structure of the cloud-topped MBL and what this implies as to the values of water vapor and temperature to be expected in recently detrained air.

The methodology for achieving the third objective involves the use of satellite retrievals of albedo (by both MODIS and GOES) to compare with in situ measurements of cloud properties and below cloud aerosol and SST gradients. The instruments to measure the in situ parameters are part of the standard instrument package on the CIRPAS Twin Otter and have been previously described.

WORK COMPLETED

Tasks associated with the first objective listed above have been largely completed for the data sets now in hand. From the RED data set, we have compiled a detailed aerosol chemical analysis similar to that previously done for both SAFARI 2000 and ACE-Asia. It is interesting to see the contrast in results between the RED data set, acquired in the background mid-Pacific, and those from the more polluted venues (reported previously). An analysis directed towards the second objective has just been completed for the CARMA-I data set and will be discussed below. No analysis dealing with the third objective has yet been completed, though one is underway.

RESULTS

Pursuing our objective of relating chemical speciation to aerosol hygroscopicity, filter data from RED were analyzed in the same fashion as that previously used in SAFARI 2000 and ACE-Asia. Results

for the filters taken from the FLIP platform are shown in Figure 1. On average, 74% of the gravimetric mass on the filters was speciated, substantially higher than could be done in more polluted locales. On the other hand, only about 5% of the mass was organic, somewhat lower than we have found previously though compatible with measurements by others in marine venues.

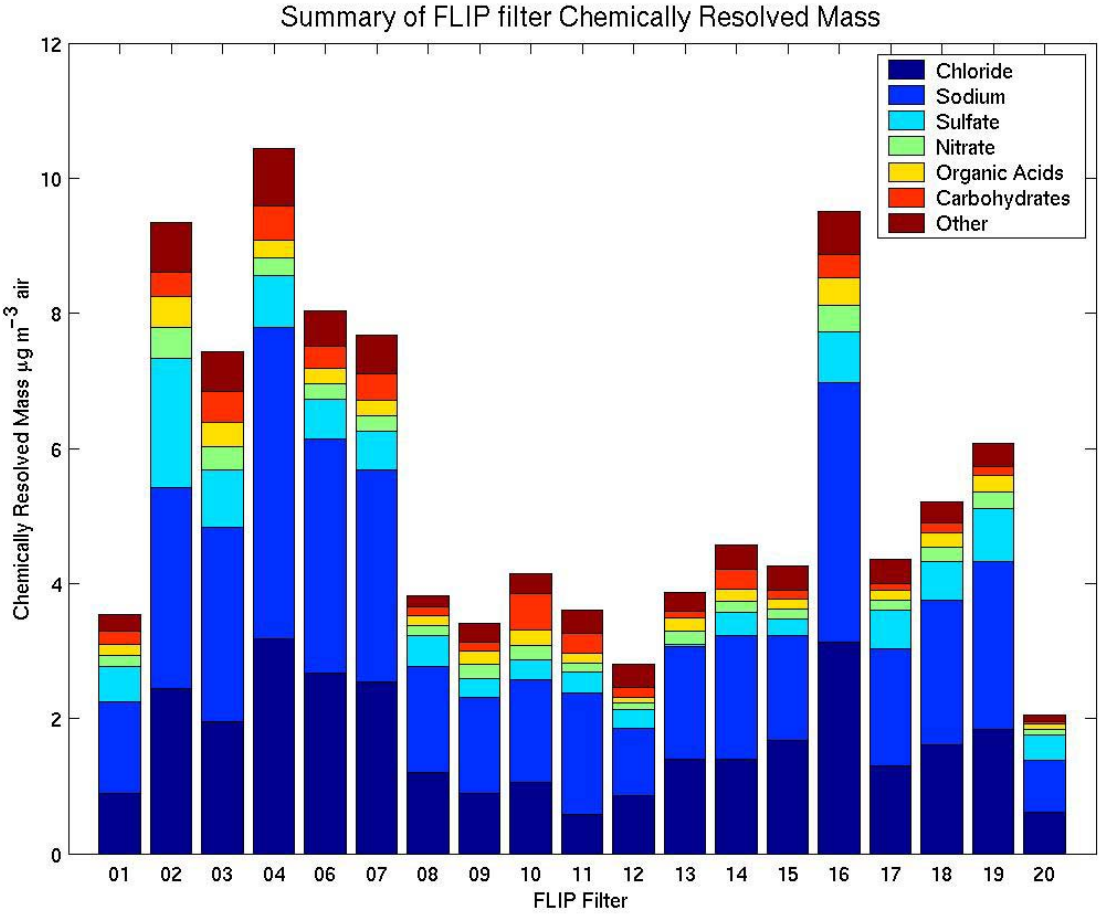

Figure 1 Summary of FLIP filter chemically resolved mass

On the basis of such chemical information as that shown in figure 1, coupled with measurements of the aerosol size distribution, the POEM model and Mie scattering code was exercised to yield hygroscopic growth curves for the aerosols analyzed during RED. Because of the lack of mass closure, assumptions had to be made as to the nature of the residual mass. Sensitivity tests were thus made with several different assumptions as to the nature of the unresolved mass. For example, one scenario explored was that in which the additional mass was a hygrophobic alkanoic acid as per the hypothesis of Ellison et al (1999). An example of model generated hydration curves for a base case (i.e., with the resolved mass alone utilized) together with curves generated with some assumptions is given in Figure 2.

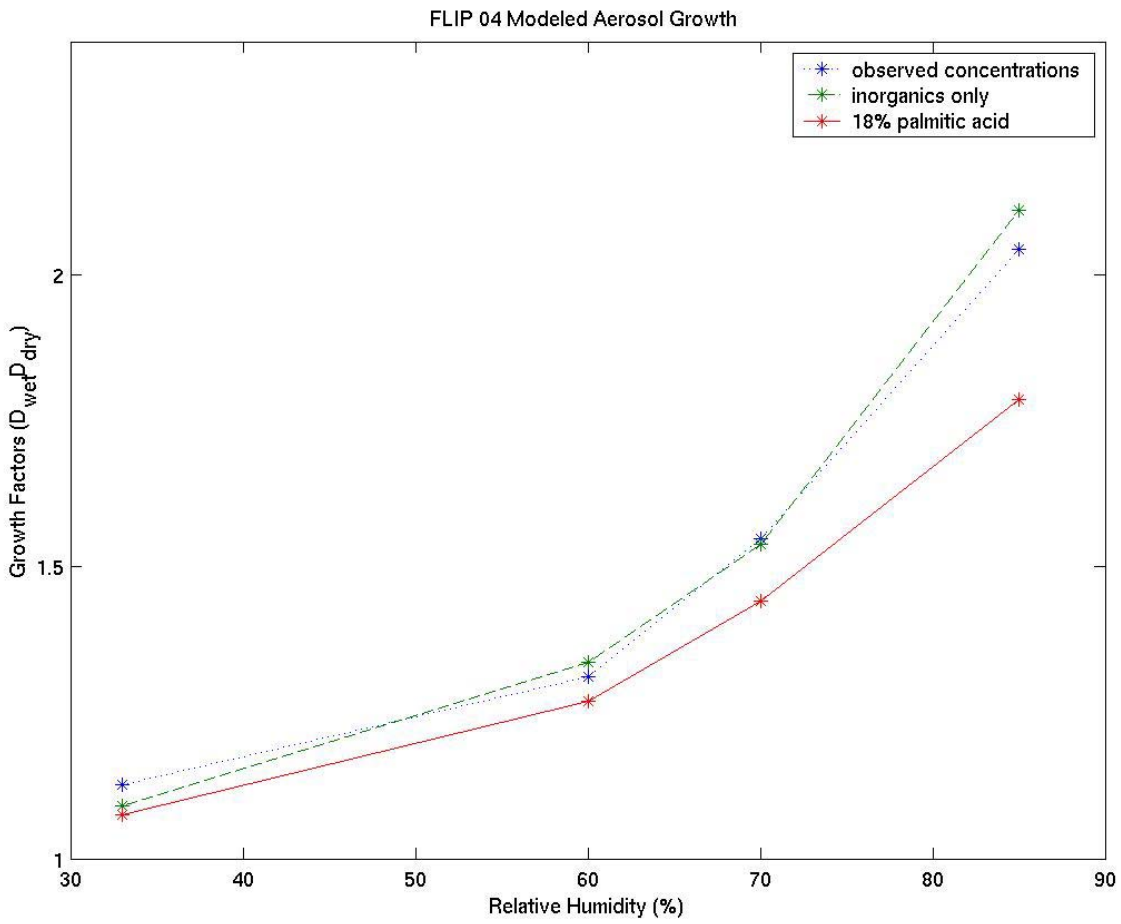

Figure 2 Modeled aerosol hydration curves for filter number four from FLIP. Examples shown are for the observed resolved mass concentrations, for only the inorganic component of the observed mass concentrations, and for the observed mass plus an additional mass of palmitic acid sufficient to achieve mass closure.

The curves show a relationship that is to be expected from their differing base compositions. When soluble organics are present, the aerosol is slightly more hygroscopic at low RH's and less so at high RH relative to the inorganic case. When the hygrophobic organic is added, essentially a surfactant, the hygroscopicity is everywhere depressed but especially at high RH, thus resulting in lower values of γ.

An analysis directed towards the second objective listed has also been completed. Utilizing fast water vapor measurements from the Lyman-α hygrometers as tracers for detraining air, we found the aerosol mass scattering efficiency in such detraining air to be significantly higher than in non-detraining air. An example of this can be seen in Figure 3.

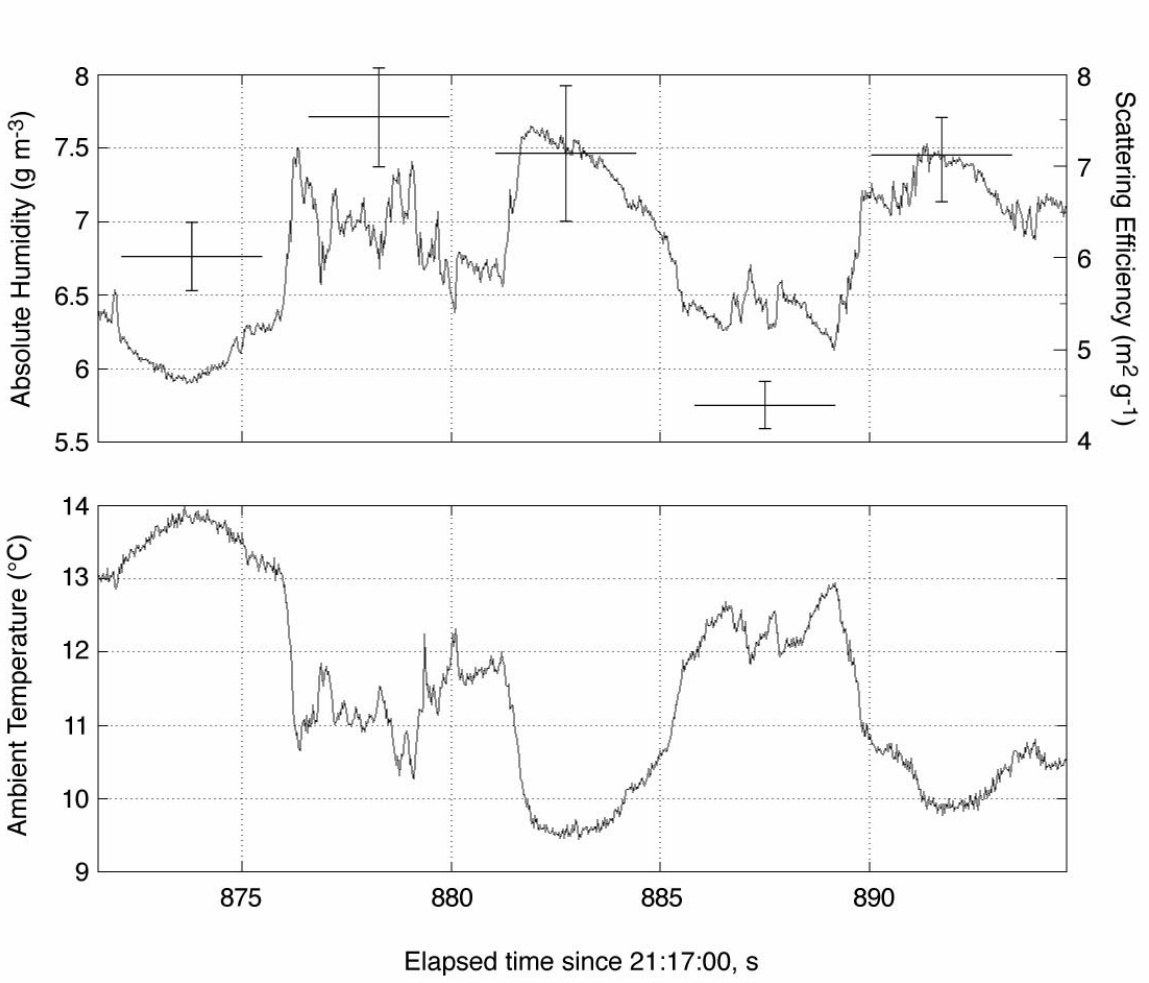

Figure 3 Time series of absolute humidity (Lyman-alpha hygrometer), temperature (Rosemount platinum wire) and aerosol mass light-scattering efficiency for a segment of the cloud-top run on August 23rd. The data rate is 40 Hz for the humidity and temperature data. The scattering efficiencies are 5 s averages of 1 Hz data.

IMPACT/APPLICATIONS

These results suggest that organics commonly play an important role in the hygroscopicity of marine aerosols. Hence, to properly assess, and predict, the impact of RH on visibility in the MBL, this organic aerosol component must be taken into account. Furthermore, the nature of the organic component, i.e., its speciation, must also be understood. Unlike the traditional inorganic sea salt model, in which the speciation is not of paramount importance since nearly all inorganic salts have roughly similar hygroscopicities, different organics can have quite different impacts upon aerosol hydration, as can be seen in Figure 2.

RELATED PROJECTS

The use of aerosol hygroscopicity and associated optical data (e.g., scattering and absorption coefficients for ambient aerosols) is of great interest to various research groups. For example, we have just completed a closure study of aerosol optical depth based on the ACE-Asia data set with Phil

Russell's group at NASA-Ames. A manuscript based on this work is now in press (JGR). Similarly, retrieval of both aerosol hygroscopicity and CCN concentration are of great interest to the remote sensing community. We are exploring the feasibility of in-situ/remote retrieval comparisons of both hygroscopicity and CCN concentration with data from both the ACE-Asia and CARMA studies.

REFERENCE

Ellison, G.B., A. F. Tuck and V. Vaida, Atmospheric processing of organic aerosols, J. Geophys. Res., 104, 11633-11641, 1999.

Ming, Y. and L. M. Russell, Predicted hygroscopic growth of sea salt aerosol, J. Geophys. Res., 106, 28259-28274, 2001.

PUBLICATIONS

Crahan, K.K., D.A. Hegg, D.S. Covert and H.H. Jonsson, Speciation of organic aerosols in the sub-tropical mid-Pacific and their relationship to light-scattering, J. Atmos. Sci., Submitted, 2003.

Gao, S., D.A. Hegg, D.S. Covert and H.H. Jonsson, Aerosol chemistry, and light-scattering and hygroscopicity budgets during outflow from East Asia, J. Atmos. Chem., In press, 2003.

Hegg, D.A., D.S. Covert, H.H. Jonsson, D. Khelif and C. A. Friehe, Observations of the impact of cloud processing on aerosol light-scattering efficiency, Tellus, In review, 2003.

Schmid, D., D.A. Hegg, J. Wang, D. Bates, J. Redemann, P.B. Russell, J.M. Livingston, H.H. Jonsson, E.J. Welton, J.H. Seinfeld, R.C. Flagan, D.S. Covert, O. Dubovik, and A. Jefferson, Column closure studies of lower troposphere aerosol and water vapor during ACE-Asia using airborne sunphotometer, airborne in-situ and ship-based lidar measurements, J. Geophys. Res., In press, 2003.

Wang, J., R.C. Flagan, J.H. Seinfeld, H.H. Jonsson, D.R. Collins, P.B. Russell, B. Schmid, J. Redemann, J.M. Livingston, S. Gao, D.A. Hegg, E.J. Welton and D. Bates, Clear-column radiative closure during ACE-Asia: Comparison of multiwavelength extinction derived from particle size and composition with results from sum photometry, J. Geophys. Res., 107, AAC 7-1 to 7-22, 2002.