

Marine Aerosols: Hygroscopicity and Aerosol-Cloud Relationships

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ONR Grant N00014-10-1-0200
October 1, 2011 – September 30, 2012

LONG-TERM GOALS

The long-term goal of this project is to predict the radiative properties of the marine atmosphere based on aerosol and cloud properties.

OBJECTIVES

The objective of the project is to improve our understanding of the role of aerosols in the Marine Boundary Layer and our ability to simulate marine stratocumulus clouds as a function of aerosol properties and the large-scale meteorology.

APPROACH

The proposed research consists of laboratory, aircraft experiment, and modeling studies that address the hygroscopic properties of aerosols and aerosol-cloud relationships in marine stratocumulus (MSc) clouds. A CIRPAS Twin Otter field experiment took place in July-August 2011 over the eastern Pacific Ocean off the coast of Monterey, CA. That experiment addressed the response of MSc to aerosol perturbations, in collaboration with Professors Bruce Albrecht of the University of Miami, Armin Sorooshian of the University of Arizona, and Lynn Russell of UCSD. We designated this experiment as the Eastern Pacific Emitted Aerosol Cloud Experiment, E-PEACE.) Evaluation of the E-PEACE data is continuing.

Report Documentation Page				Form Approved OMB No. 0704-0188	
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1. REPORT DATE 2012		2. REPORT TYPE N/A		3. DATES COVERED -	
4. TITLE AND SUBTITLE Marine Aerosols: Hygroscopicity and Aerosol-Cloud Relationships				5a. CONTRACT NUMBER	
				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) California Institute of Technology 1200 E. California Blvd., M/C 210-41 Pasadena, CA 91125				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 SAR	18. NUMBER OF PAGES 6	19a. NAME OF RESPONSIBLE PERSON
a. REPORT unclassified	b. ABSTRACT unclassified	c. THIS PAGE unclassified			

We are continuing to carry out further development and evaluation of aerosol-cloud modeling in the marine boundary layer. Much work has been carried out directed at understanding the response of marine stratocumulus to perturbations in marine aerosols, including large eddy simulation (LES) and field measurements, the latter including Twin Otter missions such as MASE I and MASE II and those involving other platforms such as DYCOMS and VOCALS, going back to the original MAST campaign. The MAST mission was the first to systematically probe the cloud properties in ship tracks, which have continued to serve as a well-defined example of marine aerosol-cloud perturbations. The Twin Otter payload has advanced considerably beyond that in the original MAST experiment, now comprising an aerosol mass spectrometer, CCN spectrometer, soot photometer, photoacoustic spectrometer, and phase doppler interferometer. It is now possible to fully characterize the aerosol physical, chemical, and optical properties. Important work remains to be done to fully understand aerosol-cloud relationships in marine stratocumulus. In addition, most of the cloud microphysical modeling done independently or in conjunction with field missions has yet to be reflected in atmospheric models. We have carried out microphysical modeling and LES studies with the goal of deriving treatments appropriate for atmospheric models.

WORK COMPLETED

A Continuous Spectral Aerosol-Droplet Microphysics Model (Lebo, Z. J. and J. H. Seinfeld, *Atmos. Chem. Phys.*, 11, 12297-12316 (2011))

A two-dimensional (2-D) continuous spectral aerosol-droplet microphysics model is presented and implemented into the Weather Research and Forecasting (WRF) model for large-eddy simulations (LES) of warm clouds. Activation and regeneration of aerosols are treated explicitly in the calculation of condensation/evaporation. The model includes a 2-D spectrum that encompasses wet aerosol particles (i.e., haze droplets), cloud droplets, and drizzle droplets in a continuous and consistent manner and allows for the explicit tracking of aerosol size within cloud droplets due to collision-coalescence. The system of differential equations describing condensation/evaporation (i.e., mass conservation and energy conservation) is solved simultaneously within each grid cell. The model is demonstrated by simulating a marine stratocumulus deck for two different aerosol loadings (100 and 500 cm^{-3}), and comparison with the more traditional microphysics modeling approaches (both 1-D bin and bulk schemes) is evaluated. The simulations suggest that in a 1-D bin microphysics scheme, without regeneration, too few particles are produced and hence the mode of the droplet size spectrum occurs at a larger size relative to the 2-D bin model results. Moreover, with regeneration, the 1-D scheme produces too many small droplets and thus shifts the mode toward smaller sizes. These large shifts in the droplet size distribution can potentially have significant effects on the efficiency of the collision-coalescence process, fall speeds, and ultimately precipitation.

Characterization and Airborne Deployment of a New Counterflow Virtual Impactor Inlet

(Shingler, T., S. Dey, A. Sorooshian, F. J. Brechtel, Z. Wang, A. Metcalf, M. Coggon, J. Mulmenstadt, L. M. Russell, H. H. Jonsson, and J. H. Seinfeld, *Atmos. Meas. Tech.*, **5**, 1259-1269 (2012))

A new counterflow virtual impactor (CVI) inlet is introduced with details of its design, laboratory characterization tests and deployment on an aircraft during the 2011 Eastern Pacific Emitted Aerosol Cloud Experiment (EPEACE). The CVI inlet addresses three key issues in previous designs; in particular, the inlet operates with: (i) negligible organic contamination; (ii) a significant sample flow rate to downstream instruments ($\sim 15 \text{ l min}^{-1}$) that reduces the need for dilution; and (iii) a high level of accessibility to the probe interior for cleaning. Wind tunnel experiments characterised the cut size of sampled droplets and the particle size-dependent transmission efficiency in various parts of the probe. For a range of counter-flow rates and air velocities, the measured cut size was between $8.7\text{--}13.1 \mu\text{m}$. The mean percentage error between cut size measurements and predictions from aerodynamic drag theory is 1.7 %. The CVI was deployed on the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter for thirty flights during E-PEACE to study aerosol-cloud-radiation interactions off the central coast of California in July and August 2011. Results are reported to assess the performance of the inlet including comparisons of particle number concentration downstream of the CVI and cloud drop number concentration measured by two independent aircraft probes. Measurements downstream of the CVI are also examined from one representative case flight coordinated with shipboard-emitted smoke that was intercepted in cloud by the Twin Otter.

Are Simulated Aerosol-Induced Effects on Deep Convective Clouds Strongly Dependent on

Saturation Adjustment? (Lebo, Z. J., H. Morrison, and J. H. Seinfeld, *Atmos. Chem. Phys. Disc.*, **12**, 10059-10114 (2012))

Three configurations of a bulk microphysics scheme in conjunction with a detailed bin scheme are implemented in the Weather Research and Forecasting (WRF) model to specifically address the role of the saturation adjustment assumption (i.e., condensing/evaporating the surplus/deficit water vapor relative to saturation in one time step) on aerosol-induced invigoration of deep convective clouds. The bulk model configurations are designed to treat cloud droplet condensation/evaporation using either saturation adjustment, as employed in most bulk models, or an explicit representation of supersaturation over a time step, as used in bin models. Results demonstrate that the use of saturation adjustment artificially enhances condensation and latent heating at low levels and limits the potential for an increase in aerosol concentration to increase buoyancy at mid to upper levels. This leads to a small weakening of the time- and domain-averaged convective mass flux ($\sim -3\%$) in polluted compared to clean conditions. In contrast, the bin model and bulk scheme with explicit prediction of

supersaturation simulate an increase in latent heating aloft and the convective updraft mass flux is weakly invigorated (~5%). The bin model also produces a large increase in domain-mean cumulative surface precipitation in polluted conditions (~18%), while all of the bulk model configurations simulate little change in precipitation. Finally, it is shown that the cold pool weakens substantially with increased aerosol loading when saturation adjustment is applied, which acts to reduce the low-level convergence and weaken the convective dynamics. With an explicit treatment of supersaturation in the bulk and bin models there is little change in cold pool strength, so that the convective response to polluted conditions is influenced more by changes in latent heating aloft. It is concluded that the use of saturation adjustment can explain differences in the response of cold pool evolution and convective dynamics with aerosol loading simulated by the bulk and bin models, but cannot explain large differences in the response of surface precipitation between these models.

Occurrence of Lower Cloud Albedo in Ship Tracks (Chen, Y. C., M. W. Christensen, L. Xue, A. Sorooshian, G. L. Stephens, R. M. Rasmussen, and J. H. Seinfeld, *Atmos. Chem. Phys.*, **12**, 8223-8235 (2012))

The concept of geoengineering by marine cloud brightening is based on seeding marine stratocumulus clouds with sub-micrometer sea-salt particles to enhance the cloud droplet number concentration and cloud albedo, thereby producing a climate cooling effect. The efficacy of this as a strategy for global cooling rests on the extent to which aerosol-perturbed marine clouds will respond with increased albedo. Ship tracks, cloud regions impacted by ship exhaust, are a well-known manifestation of the effect of aerosol injection on marine clouds. We present here an analysis of the albedo responses in ship tracks, based on in situ aircraft measurements and three years of satellite observations of 589 individual ship tracks. It is found that the sign (increase or decrease) and magnitude of the albedo response in ship tracks depends on the mesoscale cloud structure, the free tropospheric humidity, and cloud top height. In a closed cell structure (cloud cells ringed by a perimeter of clear air), nearly 30% of ship tracks exhibited a decreased albedo.

Eastern Pacific Emitted Aerosol Cloud Experiment (E-PEACE) (Russell, L. M., A. Sorooshian, J. H. Seinfeld, B. A. Albrecht, A. Nenes, L. Ahlm, Y. C. Chen, M. Coggon, J. Craven, R. C. Flagan, A. A. Frossard, H. Jonsson, E. Jung, J. Lin, A. R. Metcalf, R. Modini, J. Mulmenstadt, G. C. Roberts, T. Shingler, S. Song, E. Sumargo, Z. Wang, and A. Wonaschutz, (submitted for publication))

Aerosol-cloud-radiation interactions are widely held to be the largest single source of uncertainty in climate model projections of future radiative forcing due to increasing anthropogenic emissions. The underlying causes of this uncertainty among modeled predictions of climate are the gaps in our

fundamental understanding of cloud processes. There has been significant progress with both observations and models on addressing these important questions, but quantifying them correctly is nontrivial thus limiting our ability to represent them in global climate models. The Eastern Pacific Emitted Aerosol Cloud Experiment (E-PEACE) 2011 was a targeted aircraft campaign with embedded modeling studies, using the CIRPAS Twin Otter aircraft and the Research Vessel Point Sur in July and August 2011 off the central coast of California, with a full payload of instruments to measure particle and cloud number, mass, composition, and water uptake distributions. E-PEACE used three emitted particle sources to separate particle-induced feedbacks from dynamical variability, namely (i) shipboard smoke generated particles with 0.05-1 μm diameters (which produced tracks measured by satellite and had drop composition characteristic of organic smoke), (ii) combustion particles from container ships with 0.05-0.2 μm diameters (which were measured in a variety of conditions with droplets containing both organic and sulfate components), and (iii) aircraft-based milled salt particles with 3-5 μm diameters which showed enhanced drizzle rates in some clouds). The aircraft observations were consistent with past large eddy simulations of deeper clouds in ship tracks and aerosol-cloud parcel modeling of cloud drop number and composition, providing quantitative constraints on aerosol effects on warm cloud microphysics.

Ship Impacts on the Marine Atmosphere: Insights into the Contribution of Shipping Emissions to the Properties of Marine Aerosol and Clouds (Coggon, M. M., A. Sorooshian, Z. Wang, A. R. Metcalf, A. A. Frossard, J. J. Lin, J. S. Craven, A. Nenes, H. H. Jonsson, L. M. Russell, R. C. Flagan, and J. H. Seinfeld, *Atmos. Chem. Phys.*, **12**, 8439-8458 (2012))

We report properties of marine aerosol and clouds measured in the shipping lanes between Monterey Bay and San Francisco off the coast of Central California. Using a suite of aerosol instrumentation onboard the CIRPAS Twin Otter aircraft, these measurements represent a unique set of data contrasting the properties of clean and ship-impacted marine air masses in dry aerosol and cloud droplet residuals. Average mass and number concentrations of below-cloud aerosol of 2 $\mu\text{g m}^{-3}$ and 510 cm^{-3} are consistent with previous studies performed off the coast of California. Enhancement of vanadium and cloud droplet number concentration observed concurrently with a decrease in cloud water pH suggests that periods of high aerosol loading are primarily linked to increased ship influence. Mass spectra from a compact time-of-flight Aerodyne aerosol mass spectrometer reveal an enhancement in the fraction of organic at m/z 42 (f_{42}) and 99 (f_{99}) in ship-impacted clouds. These ions are well correlated to each other ($R^2 > 0.64$) both in and out of cloud and dominate organic mass during periods of enhanced sulfate. High-resolution mass spectral analysis of these masses from ship measurements suggests that the ions responsible for this variation were oxidized, possibly due to cloud processing. We propose that the organic fractions of these ions be used as a metric for determining the

extent to which ships impact the marine atmosphere where ($f_{42} > 0.15$; $f_{99} > 0.04$) would imply heavy influence from shipping emissions, ($0.05 < f_{42} < 0.15$; $0.01 < f_{99} < 0.04$) would imply moderate, but persistent, influences from ships, and ($f_{42} < 0.05$; $f_{99} < 0.01$) would imply clean, non-ship-influenced air.

RESULTS (Publications)

- Lebo, Z. J. and J. H. Seinfeld, A Continuous Spectral Aerosol-Droplet Microphysics Model, *Atmos. Chem. Phys.*, **11**, 12297-12316 (2011).
- Shingler, T., S. Dey, A. Sorooshian, F. J. Brechtel, Z. Wang, A. Metcalf, M. Coggon, J. Mulmenstadt, L. M. Russell, H. H. Jonsson, and J. H. Seinfeld, Characterization and Airborne Deployment of a New Counterflow Virtual Impactor Inlet, *Atmos. Meas. Tech.*, **5**, 1259-1269 (2012).
- Lebo, Z. J., H. Morrison, and J. H. Seinfeld, Are Simulated Aerosol-Induced Effects on Deep Convective Clouds Strongly Dependent on Saturation Adjustment? *Atmos. Chem. Phys. Disc.*, **12**, 10059-10114 (2012).
- Chen, Y. C., M. W. Christensen, L. Xue, A. Sorooshian, G. L. Stephens, R. M. Rasmussen, and J. H. Seinfeld, Occurrence of Lower Cloud Albedo in Ship Tracks, *Atmos. Chem. Phys.*, **12**, 8223-8235 (2012).
- Russell, L. M., A. Sorooshian, J. H. Seinfeld, B. A. Albrecht, A. Nenes, L. Ahlm, Y. C. Chen, M. Coggon, J. Craven, R. C. Flagan, A. A. Frossard, H. Jonsson, E. Jung, J. Lin, A. R. Metcalf, R. Modini, J. Mulmenstadt, G. C. Roberts, T. Shingler, S. Song, E. Sumargo, Z. Wang, and A. Wonaschutz, Eastern Pacific Emitted Aerosol Cloud Experiment (E-PEACE), *Bull. Amer. Meteorol. Soc.*, (submitted for publication).
- Coggon, M. M., A. Sorooshian, Z. Wang, A. R. Metcalf, A. A. Frossard, J. J. Lin, J. S. Craven, A. Nenes, H. H. Jonsson, L. M. Russell, R. C. Flagan, and J. H. Seinfeld, Ship Impacts on the Marine Atmosphere: Insights into the Contribution of Shipping Emissions to the Properties of Marine Aerosol and Clouds, *Atmos. Chem. Phys.*, **12**, 8439-8458 (2012).