

# **Physicochemical and Optical Characterization of Aerosol Fields from Coastal Breaking Waves**

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## **LONG-TERM GOAL**

Our long-term goal is to establish an improved understanding of the properties and factors that control the marine aerosol generation processes, their dependence on oceanic and environmental conditions and aerosol evolution in transition from pure marine to coastal environment.

## **OBJECTIVES**

Our intent is to establish a predictive capability for the size distribution of aerosol produced under various conditions, its 3-D spatial structure and associated optical effects. The focus for our study was the relation of coastal aerosol optical properties and spectral visual range. Our intent is to assess the modifications of the offshore aerosol through interactions with coastal sources and its response to various environmental factors.

## **APPROACH**

Our studies focused on breaking waves in a coastal setting through measurement of the complete size spectra from 0.007 to 15 $\mu$ m using a suite of instruments operating at both dry and ambient conditions. These have been carried out at our Bellows Air Force Station site with its 20m tower during the Shoreline Environment Aerosol Study (SEAS), during the Rough Evaporation Duct Experiment (RED) off the North coast of Oahu and from airborne platforms over the open ocean. Our instrumentation has been directed at improved understanding of aerosol production from breaking waves and to reduce uncertainties in the production and fluxes of sea-salt associated with oceanic breaking waves.

## **WORK COMPLETED (2002-2003)**

SEAS studies have led to the following publications in SEAS special issue of the Journal of Atmospheric and Oceanic Technology (v.20, #10, October 2003): A. Clarke and V. Kapustin [2]; A. Clarke et al. [3]; P. Campuzano-Jost et al. [4]; S. Masonis et al. [5].

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| 14. ABSTRACT<br><b>Our long-term goal is to establish an improved understanding of the properties and factors that control the marine aerosol generation processes, their dependence on oceanic and environmental conditions and aerosol evolution in transition from pure marine to coastal environment.</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          |                                    |                                     |                                                           |                                                     |                                 |
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A paper on our development and testing of new aerosol instrumentation – miniOPC was published in the Journal of Atmospheric and Oceanic technology [6].

From our RED measurements we found that, depending upon conditions, the degradation in EO signal could be on the order of tens of percent at a wavelength of 4  $\mu\text{m}$  (Anderson et al., submitted [7], Clarke et al., [8]).

Most recently, we re-evaluated the SEAS data to calculate oceanic production fluxes based upon our coastal breaking wave data scaled by oceanic white cap coverage (Clarke, submitted [9]).

Finally, during the past two years our ONR funding has helped support our student, Y. Shinozuka, to get his MS in early 2003. His thesis is entitled “Sea-Salt Optical properties Over the Remote Oceans: Their Vertical Profiles and Variation with wind speed”. This was based upon analysis of our aircraft data over the Pacific and Southern Oceans from prior experiments (Shinozuka et al., submitted [10]).

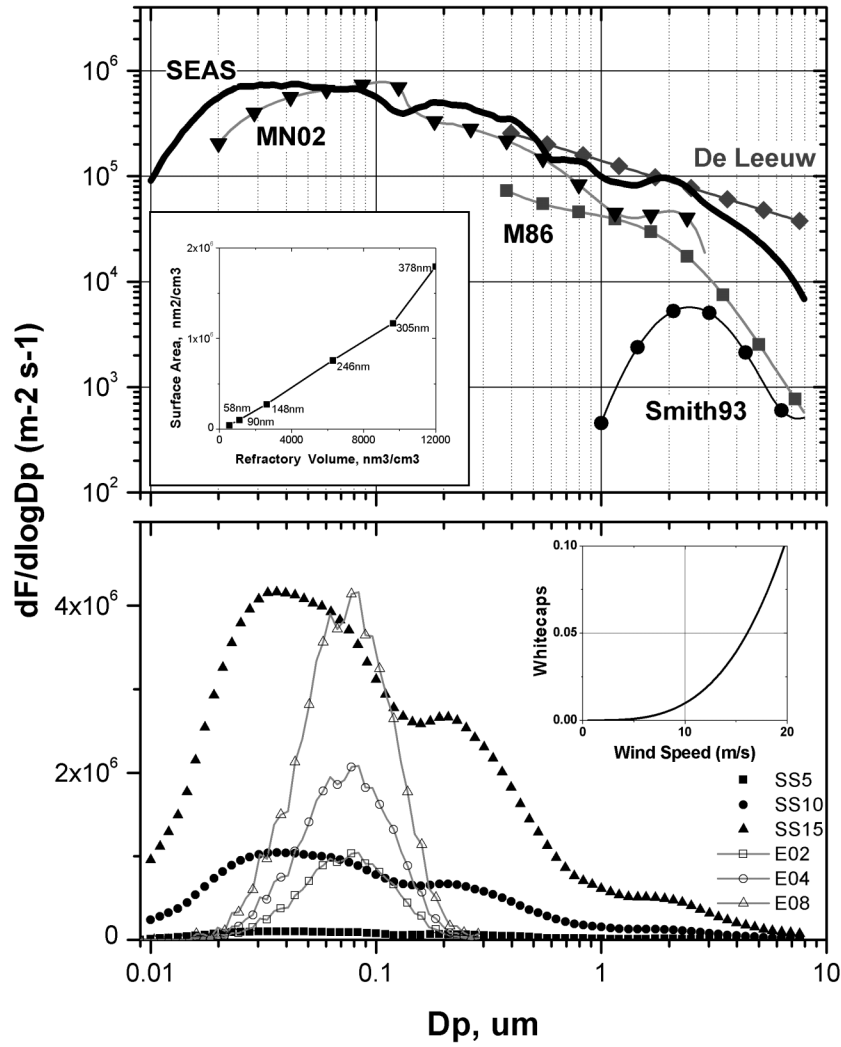
## RESULTS

### *SEAS Experiment: Marine aerosol influenced by a coastal environment and long-range transport*

SEAS (April 2000) was carried out in Hawaii on the east coast of Oahu exposed to relatively steady onshore flow. SEAS included measurements of the aerosol lidar backscatter coefficient (Masonis et al. [5]), the aerosol phase function (Lienert et al., 2003); size-resolved sodium chemistry on single particles (Campuzano-Jost et al. [4]); 3-D scanning lidar system (Porter et al., 2003), lidar signal inversion (Shifrin et al., 2003 and our measurements (Clarke et al., [1-3]).

In “The Shoreline Environment Aerosol Study (SEAS): A Context for Marine Aerosol Measurements Influenced by a Coastal Environment and Long-Range Transport”, A. Clarke and V. Kapustin [2] we discuss the role of various coastal effects at the SEAS coastal experiment site and long range transport and their influence on coastal instrumentation and measurements.

In “Sea-Salt Size Distributions from Breaking Waves: Implications for Marine Aerosol Production and Optical Extinction Measurements during SEAS”, Clarke, A. et al. [3] we present new data demonstrating sizes down to 0.01  $\mu\text{m}$  produced from breaking waves. The size distribution from these smallest sizes up through 14  $\mu\text{m}$  includes the optically effective sizes. Included is a long-term comparison of continuous size-distributions and light scattering from open-ocean and breaking wave environments that to provide an absolute calibration of concurrent lidar data. Most recently we re-evaluated the SEAS data to calculate oceanic production fluxes based upon our coastal breaking wave data scaled by oceanic white cap coverage (Clarke A. [9]). We found that particles produced from oceanic breaking waves were measured down to sizes as small as 0.01  $\mu\text{m}$ . Peak number concentrations occur at diameters of 0.02-0.06  $\mu\text{m}$  with 60% of the total number present below 0.1  $\mu\text{m}$ , about an order of magnitude smaller than previously documented for oceanic sea-salt. The measured size distribution and associated thermal volatility imply these small nuclei are sea-salt and evolve from an externally mixed to internally mixed aerosol in the marine boundary layer. Their flux from breaking bubbles (Figure 1a) and the highly nonlinear dependence of bubble production on wind speeds (insert) implies strong regional and temporal differences in their surface flux. These data have been combined with our earlier work supported by ONR at Christmas Island and with data for sulfate aerosol subsiding from the free troposphere. This provides a new recognition of the role of surface sea-salt production and entrainment on controlling the particle number and cloud condensation nuclei in the boundary layer (Figure 1b.). The figure above compares the size- resolved surface flux of sea-salt for wind speeds of 5, 10 and 15  $\text{m s}^{-1}$  and that of sulfate aerosol entrained (entrainment rates of 0.2, 0.4 and 0.8  $\text{cm s}^{-1}$ ) from the free troposphere.



**Figure 1 (a)** A comparison of observed BW source function for sea-salt determined from SEAS dry size distributions between  $0.01 \mu m$  and  $8 \mu m$  and scaled to open-ocean conditions based upon estimated white cap coverage for an assumed wind of  $9 m s^{-1}$ . Results agree with similarly scaled recent tank studies of small particle production from BW in surrogate sea-water (MN02 - Martensson et al., 2003) and with coastal studies for large sizes (de Leeuw et al., 2000). Somewhat lower values for earlier commonly referenced tank based data by Monohan et al., 1986 (M86) show fewer larger particles while Smith et al. data shows a falloff below  $2 \mu m$  not evident in other data.

Insert shows that size selected (tandem DMA) measurements on open-ocean internally mixed aerosol at CI had volumes of refractory aerosol remaining at  $350^\circ C$  that were linearly related to the surface areas of the selected unheated sizes (diameters indicated in nm on plot). Insert shows that size selected (tandem DMA) measurements on open-ocean internally mixed aerosol at CI had volumes of refractory aerosol remaining at  $350^\circ C$  that were linearly related to the surface areas of the selected unheated sizes (diameters indicated in nm on plot).

**(b)** Source flux distributions for SS (solid symbols) scaled to wind speeds of 5, 10 and  $15 m s^{-1}$  based upon our SEAS size distributions and for FT sulfate aerosol (open symbols) entrained at 0.2, 0.4 and  $0.8 cm s^{-1}$  and based upon average measured distributions over the tropics/subtropics between 1 and 2 km altitude. Insert shows whitecap coverage vs. wind speed.

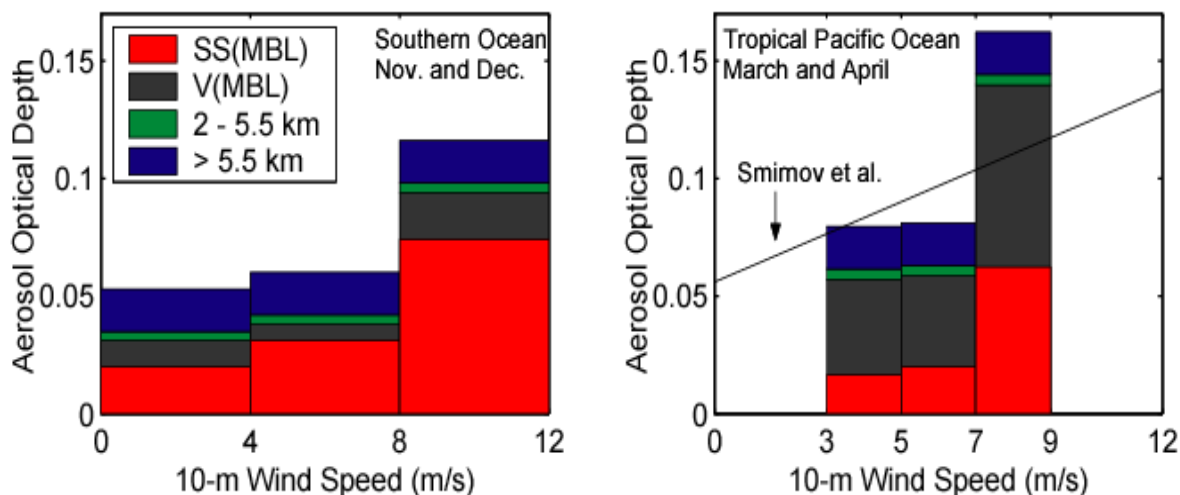
These sources are linked to their relative role as condensation nuclei and cloud condensation nuclei, CCN. The implications of this SS surface flux extend to not only optical transmission issues but also climatological consideration including influences on cloud optical properties in remote oceanic regions.

***RED experiment: The Contribution of Coastal Aerosol From Breaking Waves to Visible and IR Light-Extinction Over 10 Km Path***

The Rough Evaporation Duct (RED) experiment was off the NE coast of Oahu, Hawaii in August-September of 2001. A 10km electro-optic (EO) path between the R/V FLIP and a receiver on the coast passed over a near shore region of frequent breaking waves about 500m in front of the detector. Marine aerosol produced from these breaking waves varied in intensity of production and their influence on the EO signal depending upon environmental factors including wind direction. The aerosol plumes increased measured aerosol extinction by a factor of about 20 relative to open ocean values (Clarke, [8]). The resulting estimated IR extinction by aerosol from the surf zone alone could approach the values for open-ocean extinction over EO path to FLIP and explain much of the reduced IR transmittance.

***Sea-Salt Optical properties Over the Remote Oceans: Their Vertical Profiles and Variation with wind speed***

During the past two years we re-evaluated our aircraft data from prior experiments over the Pacific and Southern Oceans. The focus was on the vertical distribution of coarse sea-salt (SS) and fine particle volatile “sulfate” (V) and their optical properties (Shinozuka et al., submitted [10]), as in Fig.2 below.



***Figure 2. Aerosol column optical depth from measured aerosol size and volatility averaged over ACE 1 (left) and PEMT-B (right) observations for the 10-m wind speed categories. The legend reads (from top) sea-salt in the MBL, volatile particles in the MBL, aerosols over 2 – 5.5 km and aerosols over 5.5 km – top of atmosphere. Total column AOD observed by AERONET (Midway) given by Smirnov et al. [2003] is also plotted in the right panel against surface wind speed averaged within 24 hours prior the instantaneous optical measurement.***

## IMPACT/APPLICATION

Our mini-OPC was demonstrated [6] to be an effective new tool for aerosol measurements. Our observations of sub-100nm sea-salt aerosol [2,3,9] will lead to revised understanding of sea-salt production. SEAS [2-5] highlighted new approaches for calibration of lidars and to derive aerosol extinction values. RED aerosol data [7, 8] allowed estimate the effect of coastal aerosol from breaking waves on visible and IR EO propagation.

## RELATED PROJECTS

Our SEAS measurements are directly linked with Dr. S. K. Sharma's Lidar project ONR #N000149610317, Dr. K. S. Shifrin ONR project # N000149810773 and Dr. J. N. Porter's radiation project NASA-NAG-56340. Our RED coastal measurements are directly linked with Dr. K.D. Anderson Electromagnetic Propagation project ONR #N0001401WX20195 and other RED related projects. The mini-OPC tests contributed to and were also partially supported by activities under our NASA-TRACE-P funding (NCC-1-416).

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