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1. INTRODUCTION

Aerosols have a large effect on climate and weather. They modulate the surface irradiance in clear sky conditions, but also modify the radiative properties of clouds. To assess the consequences of reduced surface irradiation due to aerosols for weather forecasting and climate, data on aerosol radiative effects and their spatial and temporal variations are required over a large area. The necessary 4-dimensional distribution of aerosol properties can be provided by integrating ground-based, airborne and satellite observations through data assimilation with advanced models. The use of satellite data to constrain Global Climate Models (GCM) or chemical transport models (CTM) has been demonstrated [Buitjes et al., 2001; Verver et al., 2002]. This leads to a new era of high potential for providing continuous availability of high quality data for use in, e.g., weather forecasting, climate studies and air quality assessment.

With 70% of the Earth covered by oceans, the occurrence of sea salt, produced primarily by breaking wind waves, is an important factor. Sea salt aerosol production has been observed in the size range from 20nm [Mårtensson et al., 2002] to over 100 μ m [De Leeuw, 1993]. Sea salt is the dominant submicrometer scatterer in most ocean regions and dominates the marine boundary layer particulate mass concentration in remote oceanic regions, with a significant fraction occurring in the submicrometer size range [IPCC, 2001]. Sea salt contributes 44% to the global aerosol optical depth. Estimates for top-of-atmosphere, global-annual radiative forcing due to sea salt are -1.51 and -5.03 Wm^{-2} for low and high emission values, respectively [IPCC, 2001]. At lower levels in the atmosphere, sea spray particles can dominate atmospheric propagation at visible and infrared wavelengths. The main contribution to the indirect aerosol effect (IAE), the most uncertain forcing mechanism in the prediction of climate change, comes from marine stratocumulus clouds. Boers *et al.* [1998] report an example of IAE related to changes in the

distributions of natural CCN over the ocean, and O'Dowd et al. [1999a, b] have demonstrated from aircraft observations that sea spray particles can play a significant role in marine stratocumulus microphysics and chemistry. In addition, sea spray particles can provide a significant sink for natural and anthropogenic trace gases [O'Dowd et al., 2000].

These examples show the importance of an accurate knowledge of sea spray aerosol emissions and resulting concentrations. Global circulation models (GCM) and chemistry transport models (CTM) need to incorporate source functions for all significant aerosol (and aerosol precursor) emissions, plus treatments of heterogeneous chemistry to deal with the transformations and removal of anthropogenic pollutant gases.

2. SEA SPRAY SOURCE FUNCTIONS

Estimates of the surface flux of sea spray aerosols, the sea spray source function, vary by six orders of magnitude [Andreas, 1998]. However, Andreas argues that most of the source functions can be disregarded, which constrain the source function estimates to within about one order of magnitude. Monahan et al. [1986] (M86) is considered representative for the bubble-mediated part of the source function, and Smith et al. [1993] (S93) is derived from field experiments using the balance between production and removal and thus includes all sea spray droplets. M86 and S93 are most widely used in chemical transport models.

Experimental efforts since 1998 are reviewed in Schulz et al. [2002]. They vary from airborne experiments to determine the aerosol loading of the marine boundary layer with increasing fetch [Reid et al., 2000] to direct eddy correlation measurements [Nilsson et al., 2001] and yield similar results as M86 and S93. For example, Figure 1 shows that the sea spray aerosol source function derived with the coastal aerosol transport model CAT [Vignati et al., 2001] from measurements over the North Atlantic by

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O'Dowd et al. [1997] compares favorably with M86 and S93.

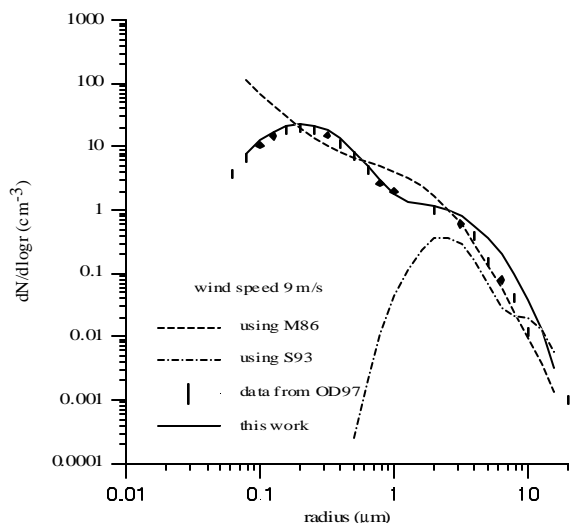


Figure 1. Comparison of sea spray source functions derived by Vignati et al. [2001] with CAT, M86 and S93 with experimental data from O'Dowd et al. [1997] (from Vignati et al. [2001]).

However, Gong et al. [1997] show that the aerosol concentrations in the marine atmosphere vary by one order of magnitude. Hence the methods applied by Vignati et al. [2001] or Smith et al. [1993] are expected to vary with the location in a similar manner. This is in good agreement with the current uncertainty in the sea spray source functions of one order of magnitude discussed above.

Such measurements are usually made in specific areas, but differences in oceanographic or meteorological parameters other than wind speed or the friction velocity are not accounted for. M86 relies on the whitecap cover, which has been parameterized in terms of water temperature and atmospheric stability. The question arises which parameters need to be accounted for to further constrain a universal source function with such detail that effects of sea spray aerosol on, e.g. climate, can be accurately estimated. Our efforts to study the sea spray source function are aimed at a combination of various approaches, including direct eddy correlation measurements, bubble measurements, aerosol concentration profiles near the air-sea interface, and modeling. In this contribution we address bubble-mediated production of aerosols, and in particular the variability of bubble concentrations.

3. THE RED EXPERIMENT

Aerosol and bubble measurements were undertaken during the RED (Rough Evaporation Duct) experiment in August/September 2001. RED took place at the Pacific at six miles from the northeast shore of Oahu, Hawaii. The measurements described here were made from the moored Floating Marine

Physical Laboratory's Research Platform Floating Instrument Platform (FLIP), see Figure 2. Aerosol size distributions were measured with optical particle counters. A volatility system was used to characterize the aerosol components, in particular to obtain information on the contribution of sea spray to the aerosol spectra. In combination with measured bubble size distributions, this information is used to study the bubble-mediated production of sea spray particles. An aerosol transport model is used to relate the derived source functions and measured concentrations. Backward trajectories show that the air masses encountered during the measurements had been over the ocean for at least three days, thus reducing the influence of continental sources.

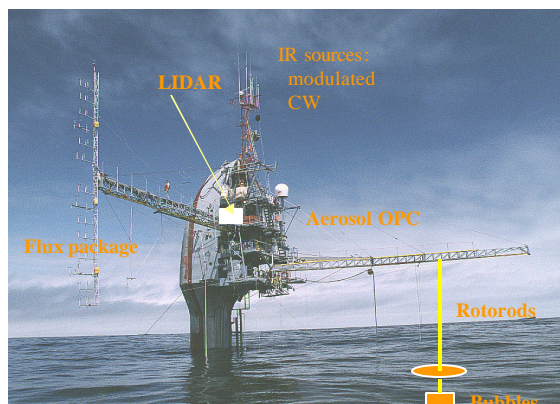


Figure 2. FLIP

4. AEROSOL CONCENTRATIONS

Aerosol time series measured on FLIP during the 3-weeks deployment off Oahu (HI, USA), Figure 3, show rather constant concentrations with a modulation indicating diurnal effects, not related to RH effects. In addition, three periods can be identified with increased concentrations. The first period is associated with pollution induced by Hawaii, as indicated by air mass trajectories. The enhanced concentrations are mainly visible in the small (sub-micron) particle range. The second period with enhanced aerosol concentrations, around 30 August, was associated with elevated wind speeds resulting in more whitecapping and thus production of sea spray aerosol. The latter is the subject of this study.

5. AEROSOL CONCENTRATION PROFILES

Because sea spray is produced at the sea surface, the concentrations are commonly expected to show distinct near-surface gradients. However, numerous measurements of aerosol profiles in the atmospheric surface layer show that usually such gradients do not exist (e.g., De Leeuw [1993]). Also during RED no clear gradients were detected. In view of the relatively low wind speeds encountered during RED (12 ms^{-1} max.) bubble-mediated sea spray

aerosol production will have been the controlling mechanism.

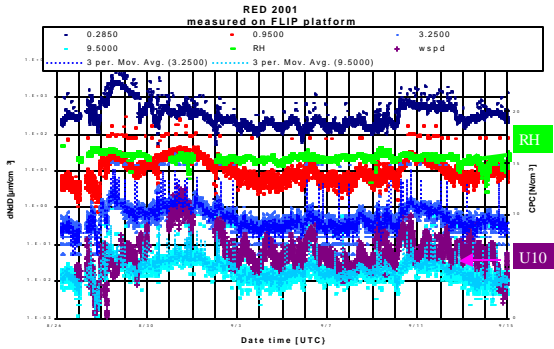


Figure 3. Time series of concentrations of aerosol particles with diameters of 0.28, 0.95, 3.2 and 9.5 μm, relative humidity and wind speed.

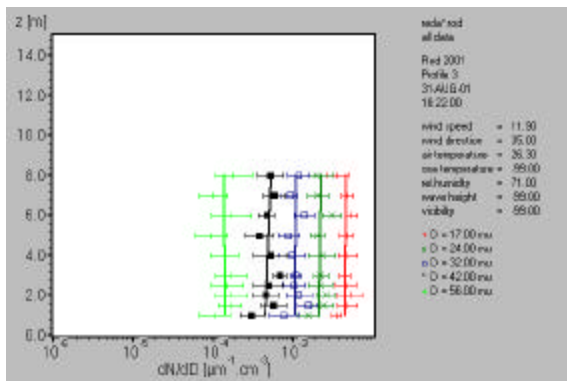


Figure 4. Particle concentration profiles measured in a wind speed of 11.9 ms⁻¹.

6. BUBBLES

Bubble spectra in the diameter range of 30-1000 μm were measured with a calibrated optical bubble measuring system (Mini-BMS) (Leifer et al., 2002). Bubble spectra were measured every three hours, as 15-minute averages, in wind speeds varying from 6 to 12 ms⁻¹.

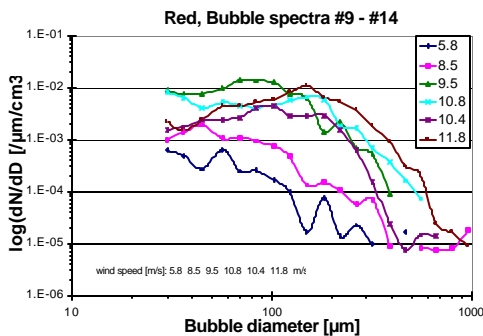


Figure 5. Bubble spectra measured from FLIP in wind speeds varying from 6 to 12 ms⁻¹.

Over this range of wind speeds, the bubble concentrations vary by about 2 orders of magnitude, depending on the bubble size. It is noted that these spectra are 'background spectra', i.e. representative for the average bubble concentration. Laboratory measurements show that the concentrations immediately after wave breaking are much higher, and the evolution of the bubble spectra depends on parameters such as fetch, bubble size, penetrations depth and plume type [De Leeuw et al., 2001; Leifer et al., 2001].

The wind speed dependence of the oceanic 'background' bubble concentrations at various locations has been presented before, cf. De Leeuw and Cohen [2001] for an overview. The bubble concentrations measured during RED show an explicit wind speed dependence

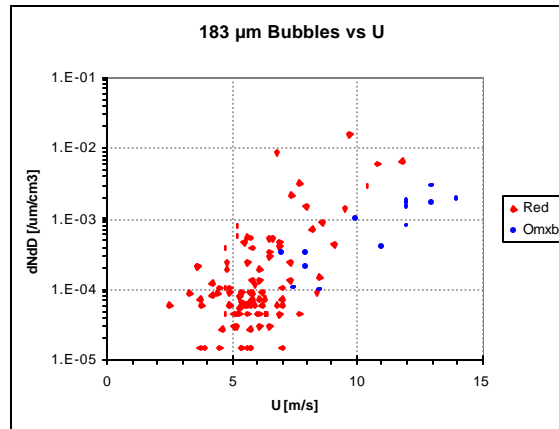


Figure 6. Wind speed dependence of the concentrations of bubbles measured from FLIP near Hawaii in a water temperature of 28°C, and in the North Atlantic in a water temperature of 12°C.

Fitting an exponential function:

$$\log\left(\frac{dB}{dD}\right) = a + bu_{10}$$

yields the slopes b describing the variation of the bubble concentrations with wind speed. Figure 7 clearly shows the strong variation of the wind speed dependence with the bubble size.

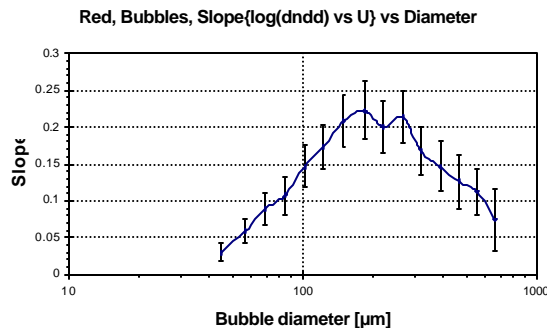


Figure 7. Variation of the wind speed dependence of bubble concentration with bubble size.

Furthermore, the concentrations and their wind speed dependence depend on the water temperature. The RED experiments took place in water with a temperature of 28°C. In Figure 6 also OMEX data are plotted which were measured in the North Atlantic with a temperature of about 12°C. Figure 8 shows an attempt to relate the variation of the concentrations with the water temperature, for wind speeds of 8-10 ms⁻¹, with data from 8 experiments in which the water temperature varied between 9 and 28 °C.

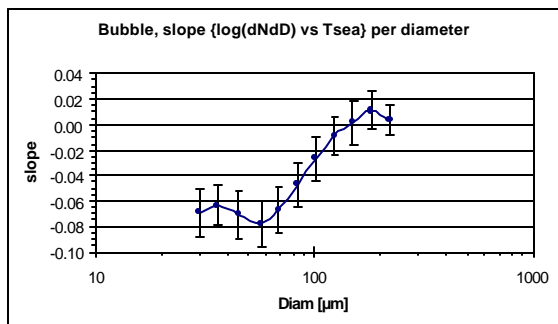


Figure 8. Variation of the water temperature dependence of the bubble concentrations with bubble size, for a wind speed of 8-10 ms⁻¹.

7. DISCUSSION AND CONCLUSIONS

- Bubble concentrations are strongly variable due to a variety of environmental factors; indicated here are effects of wind speed and water temperature:
 - Increase with wind speed;
 - Decrease with sea temperature when small, increase with sea temperature when large;
 - Spectral shape changes with wind speed and sea temperature.
 Other effects are surfactants, salinity, viscosity, Langmuir circulations, turbulence, wave breaking characteristics influenced by fetch and swell, water saturation, atmospheric thermal stratification, etc.
- Wave breaking is intermittent, and thus the bubble concentrations and subsequent sea spray aerosol production vary in time.
- Aerosol gradients depend on atmospheric transport and transformation and removal processes.

All these processes carry uncertainties. With the rather simple methods to derive the aerosol source function, either from the number of aerosols produced per unit whitecap, or from the balance between production and removal, it cannot be expected that a single universal sea spray source function can be obtained in terms of only few parameters. Yet current formulations for the sea spray source function are converging to within less than one order of magnitude.

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