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Evolution of the vertical profile and flux of large sea-salt particles in a coastal zone

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Abstract. In the vicinity of the North Carolina Outer Banks we observed both steady onshore flow conditions and a continental air mass transition into a marine boundary layer. Using the CIRPAS Twin Otter aircraft, we measured changes in the column burden of sea salt as the air mass was advected out to sea. We also measured the flux of whitecap-generated sea-salt particles in neutrally stable atmosphere at wind speeds of 4, 8, and 12 m s^{-1}. Production of salt particles as small as 0.27 μm in diameter was observed. Furthermore, we measured salt particle size distributions at various wind speeds during along shore wind and near steady state conditions. Using these measurements as a frame of reference, we discuss the very large differences in the reported size and flux of sea salt presented in the literature. The disagreement in reported salt fluxes is larger for smaller-sized particles (almost an order of magnitude) and is most likely due to assumptions made when the fluxes were computed, especially the particle dry deposition velocity and air mass history. However, for giant salt particles with short atmospheric lifetimes ($>\sim$10 μm in diameter), there is general agreement between fluxes and size distributions measured in this study and previous ones. Reported salt particle size distributions in the literature also vary considerably under similar steady wind and stability conditions. From these and our results it is clear that no more than half of the variance in salt particle concentration can be explained by wind speed alone, suggesting that the idea of "steady state" in the marine boundary layer rarely exists at midlatitudes.

1. Introduction

Measurements of sea-salt and sea-spray fluxes reported in the literature vary by several orders of magnitude [*Andreas*, 1998]. It is troubling that such variations exist, especially when one considers the importance whitecap-generated particles may play in cloud microphysics [*Johnson*, 1982; *Bower and Choularton*, 1994; *Hegg*, 1999; *Feingold et al.*, 1999]. Modeling studies suggest that the presence of large and giant sea-salt particles at cloud base in concentrations as low as 1 L^{-1} may influence marine cloud reflectivity and longevity [e.g., *Johnson*, 1982; *Feingold et al.*, 1999]. Such sensitivity to coarse mode particles can cause large uncertainties when cloud fields are modeled. The important role of coarse mode sea-salt particles in OH chemistry has also been demonstrated [e.g., *Finlayson-Pitts et al.*, 1999].

Some of the variability in salt particle fluxes and size distributions is due to variations in wind speed and wave height [*Blanchard et al.*, 1984; *Hoppel et al.*, 1989; *Fitzgerald*, 1991; *Porter and Clarke*, 1997]. The vertical distribution of large sea-salt particles is dependent on stability and convection related to air/sea temperature differences [*Fairall et al.*, 1983; *Blanchard et al.*, 1984; *Exton et al.*, 1986]. For similar meteorological conditions, however, measured salt particle size distributions vary considerably in the literature (see *Fitzgerald* [1991] and *Porter and Clarke* [1997] for comparisons). Compounding this variability in nature are measurement errors associated with coarse mode aerosol characterization. Until the flux, size, and vertical distribution of sea salt can be sufficiently parameterized, results from marine cloud and chemistry models will likely have high uncertainties.

Collectively, the uncertainties in salt particle size and flux parameterizations may be unacceptably high for use in modeling studies. By combining the *Monahan et al.* [1986] and *Smith et al.* [1993] data sets through logical deduction, *Andreas* [1998] attempted to reduce this uncertainty in the flux parameterizations and derived a new flux parameterization for wind speeds up to 32 m s^{-1}. The purpose of this paper is to test this and other sea-salt flux and size distribution parameterizations and to attempt to shed light on the true uncertainties of the state of the art. This is done in the context of data from several research flights measuring the sea salt in a coastal zone during both steady and nonsteady state conditions.

The production, size distribution, and vertical distribution of sea-salt particles produced by surf and whitecaps in a coastal zone were studied as part of the Electro-Optical Propagation Assessment in Coastal Environments (EOPACE) 1999 winter field campaign. An intensive operations period was performed from February 22 to March 12, 1999, in the vicinity of the Army Corps of Engineers research pier at Duck, North Carolina (Outer Banks). As part of this study, for 8 days (35 hours) we flew the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) UV-18A Twin Otter in the Outer Banks regions studying salt particle microphysics.

In this paper, we first present size distributions of sea salt measured during steady onshore and alongshore flow condi-

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tions. We examine these "steady state" conditions of the marine boundary layer near the shore and several hundred kilometers out to sea. Second, we present the vertical distribution of coarse mode sea-salt particles for the 3 days when we had offshore flow with similar air/sea temperatures and atmospheric stability but varying wind speed (u = 4, 8, and 12 m s^{-1}). By performing a box model calculation with these data we derive directly the sea-salt flux at 30 m.

In the context of the results of this study we discuss several parameterizations that are frequently used. We examine how well-published flux and size parameterizations compare to the present study and among themselves and attempt to partially explain the large differences found.

2. Study Design

For this study the CIRPAS UV-18A Twin Otter carried basic meteorological instrumentation (Rosemount temperature, Edge Tech dew point, Vaisala relative humidity and temperature, static pressure) and two aerosol particle probes Particle Measuring System, Inc., Forward Scattering Spectrometer Probe (PMS FSSP)-100 and Passive Cavity Aerosol Spectrometer Probe (PCASP)-100×). Nominally, the FSSP and PCASP measure particle size from 1 to 26 μm and 0.1 to 3 μm in diameter, respectively. However, because of uncertainties in detection thresholds, data from the first and last channels for these probes are not used. This reduced the probe size ranges to 2–24 μm and 0.11–2.5 μm for the FSSP and PCASP, respectively. Immediately prior to EOPACE, the PCASP and FSSP were overhauled at Droplet Measurement Technologies, Inc., in Boulder, Colorado, equipped with new data systems and calibrated. Probes were postcalibrated at CIRPAS.

For the overlap region of the two probes (2–2.5 μm) the probes compared fairly well, with the FSSP being systematically ~25% lower in concentration than the PCASP. A portion of this difference can be explained by noting that the PCASP and FSSP measure partially dried and ambient particles, respectively. In some calculations and figures it was necessary for comparison reasons to present a humidified version of the PCASP spectrum to ambient relative humidity (RH) using the marine aerosol humidity parameterization of *Gerber* [1985] for sizes down to 0.3 μm (salt dominated). Below 0.3 μm we used the hygroscopicity curve found by *Kotchenruther et al.* [1999] for East Coast aerosols. Generally, however, size spectra presented in this manuscript for sizes <2 μm are for dried aerosol particles. We do this to highlight the submicron aerosol production. If an ambient RH growth factor was used, uncertainties in that parameterization would create difficulties in distinguishing between "new" particles and background particles that grew into the larger bins due to the increasing RH. Thus there must be some caution in interpreting some of the size spectra presented. We specify in the text and captions which RH is used for submicron aerosols.

During the EOPACE/ Duck 1999 field study, the Twin Otter flew eight flights for 35 flight hours in the North Carolina Outer Banks region (see Figure 1). Two basic flight plans were performed: coastal and open ocean. Twenty-eight flight hours used the coastal flight plan to observe a continental air mass as it was advected out to sea. Flight legs, flown at 1500, 600, 300, 150, 60, and 30 m, started at the Army Corps of Engineers pier (latitude 36.18°N, longitude 75.75°W) and continued off shore for ~50 km in a direction normal to the shoreline (~78° east of north). Gradual vertical profiles from 1500 m to the surface were also performed at the shoreline and 50 km out (the continental planetary boundary layer (PBL) was typically ~1000 m thick). In order to get statistically significant samples near the shore, the Twin Otter also flew parallel to the shoreline 4 km up and down from the pier at distances of −3, 0, 1, and 3 km from the shore. This pattern was performed at various elevations between 30 and 300 m, depending on conditions. Approximately 2.5 hours were required to complete the flight legs and vertical profiles. To ensure that there were no significant changes in the boundary layer state during the flight, the 30- and 60-m flight legs were flown repeatedly.

On two occasions, the Twin Otter measured marine aerosols using the open ocean flight plan. It was flown to the Gulf Stream boundary, ~200 km from shore. For this flight plan, vertical profiles were performed with 30-km-long legs at 1500-, 600-, 100-, and 30-m levels across the Gulf Stream boundary.

3. Observations

Table 1 summarizes the research flights, listing the surface (3 m) wind speed and direction taken at the end of the Army Corps pier, as well as the flight date and the pattern flown. Within 50 km of shore, sea surface temperatures (SST) were ~8°C, and air temperatures varied from 6 to 12°C. This temperature difference corresponds to a neutral to slightly stable stability classification [*Hsu*, 1992]. For the two flights over the open ocean, SSTs increased rapidly as we passed over the Gulf Stream to 15–20°C, creating more unstable conditions.

Near-shore aerosol concentrations for the eight study flights are presented in Plate 1. For the 4 days of alongshore flow (February 26, February 27, March 11, and March 12, in Plates 1a, 1b, 1g, and 1h, respectively) the significant influence of surf line generated sea salt is clearly evident. For the 1 day of onshore flow, March 8 (Plate 1f), a fairly steady and well-mixed aerosol distribution was present. For the 3 days of offshore flow (March 1, 2, and 4 in Plates 1c, 1d, and 1e, respectively), salt particle concentrations are very low, with almost no impact from the surf line. In sections 3.1–3.3 we discuss these three flow regimes separately.

3.1. Steady State

On several occasions the CIRPAS Twin Otter measured marine aerosols that were in near steady state. National Centers for Environmental Prediction (NCEP) back trajectories suggested that the air mass had been over the water for at least 8 hours (highest wind speed case) and at least 12 hours for all others. This includes the March 8 case depicted in Plate 1f, the March 12 case (~50 km out to sea), and the 2 days the Twin Otter flew several hundred kilometers out over the ocean. While this is not enough time for submicron aerosols to come to equilibrium, coarse mode salt particles near the surface may be assumed to be close to equilibrium. (For example, *Fairall et al.* [1983] estimated that particles larger than 10 μm in diameter come into equilibrium after ~11 hours at 400 meters altitude. Near the surface it is probably faster. See section 5 for elaboration.) The 30–100 m number and volume distributions of coarse mode sea-salt particles for these "equilibrium" conditions are given in Figures 2a and 2b, respectively. Relative humidity for these cases was 70–80%. For the 5 and 14 m s^{-1} cases, measurements were taken over the open ocean at ~35.5°N, 75.8°W. For the 8 and 11 m s^{-1} cases, measurements were taken during periods of onshore flow at ~50 km away at the end of our vertical profiles for the coastal flight plan.

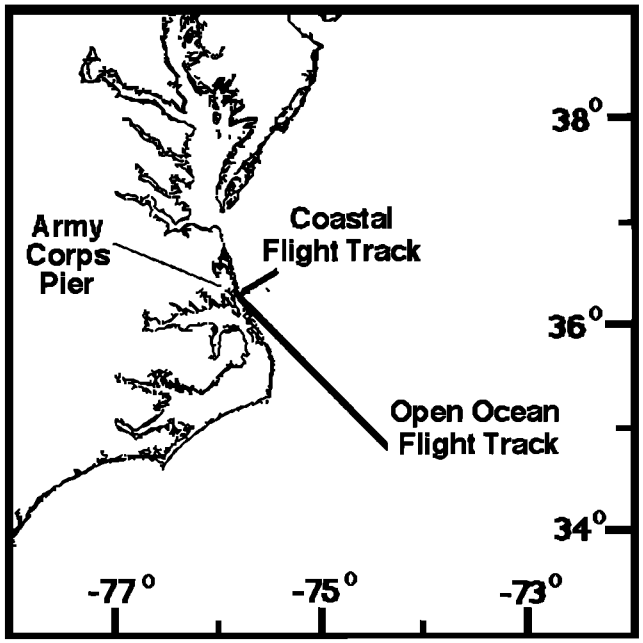

Figure 1. Map of EOPACE/Duck 1999 area of operations. Two principal flight tracks shown as bold lines.

Figure 2 clearly shows the well-known aerosol concentration versus wind speed relationship for these cases. Higher wind speeds produce more particles with a $\sim u^2$ to u^3 dependence depending on particle size. The ambient salt particle volume median diameter (VMD) varies between 8 and 10 μm. The volume distribution also has the familiar shape of larger particle sizes for higher wind speeds. At ~8–10 μm the VMD for this bubble-produced salt mode is about the same as those measured by other investigators using similar methods [e.g., *Sievering et al.*, 1987; *Hoppel et al.*, 1989]. For the 11–14 m s^{-1} cases we can begin to observe spume production of salt particles at diameters >14 μm.

Figure 3 shows the vertical distribution of coarse mode sea salt for three steady state cases. All these steady state cases are close to those found by *Blanchard et al.* [1984] measured under similar stability conditions. Figure 3a shows data from the 5 m s^{-1} open ocean case (March 2). In this case, where stability was slightly less than neutral, we see that while the sea-salt concentration is low, it is nevertheless well mixed up to the top of the 900-m boundary layer. Also, larger particles, $d_p > 7$ μm, appear well mixed to the top of the boundary layer (the apparent variability is likely due to poor counting statistics; here we counted ~1 particle every 30 s). Because of the low wind speed the concentration of particles larger than 15 μm (not shown) was below statistically significant levels, with <1 particle counted every several minutes. The vertical profile of these particles could not be distinguished from noise.

Figure 3b shows the vertical distribution of particles 50 km off shore, at 11 m s^{-1} wind speed, and neutral stability (March 8). Like the case shown in Figure 3a, coarse particles were well mixed from 30 m up to the top of the boundary layer. Even the largest particles ($15 < d_p < 24$ μm) were well mixed (the apparent variations in the vertical distribution of these largest particles in the lowest 400 m are due to poor counting statistics).

Finally, Figure 3c shows a slightly unstable case (March 11) over the Gulf Stream for wind speeds of 14 m s^{-1}. On this day, air temperature was 3°C, and water temperature was ~18°C. Steam trails in the lowest few meters above the waves were visible starting at the cold/warm water boundary. Also, a sharp stratocumulus cloud deck formed at the warm water boundary in the 600–1100 m altitude range. In this case, not only is the marine boundary layer well mixed, but there is also an apparent increase in the number of particles with height. Giant particle concentrations just below cloud base were ~2–3 L^{-1}. The apparent increases in particle concentration with height are probably related to the increase in relative humidity with height up to cloud base. Such an increase causes particles to grow and shifts the size distribution toward larger sizes, thus generally increasing the number of particles in the various size bins. Above 600 m the Twin Otter flew between clouds, and cloud-contaminated data were removed. However, some cloud droplets may have survived the screening process, resulting in the increase in particle concentration at the cloud altitudes.

3.2. Along-Shore Flow

For the 4 days when winds traveled along the shoreline, the impact of surf-generated salt particles (as opposed to whitecap-generated) was clearly visible. In these cases, salt particles from the surf significantly enriched particle concentrations out to 5–8 km. This is even true for the case of March 11, when there were significant whitecaps offshore.

Size distributions of sea salt generated by the surf line are very similar to those presented in section 3.1 (see Figure 4). Particle count and volume median diameters ranged from 1 to 4 μm and 6 to 10 μm, respectively. Geometric standard deviations (σ_g) ranged from approximately 1.8 to 2.0. Even at specific wind speeds, VMDs for the along shore flow and steady conditions are the same: ~7 μm for 5 m s^{-1}, 9 μm for 11 m s^{-1}, etc.

The vertical extent of the shoreline plumes was remarkably low, typically only a few hundred meters. To a large part, this

Table 1. Summary of Flights[a]

Date	Surface Wind at Pier	Near-Shore Flights	Open Ocean
Feb. 26, 1999	NW at 5 m s^{-1}	X	
Feb. 27, 1999	south at 3 m s^{-1}	X	
March 1, 1999	WSW at 8 m s^{-1}	X	
March 2, 1999	WSW at 4 m s^{-1}	X	X
March 4, 1999	WSW at 12 m s^{-1}	X	
March 8, 1999	north at 11 m s^{-1}	X	
March 11, 1999	NNW at 10 m s^{-1}	X	X
March 12, 1999	NNW at 7 m s^{-1}	X	

[a]Date, surface wind speed, and direction at the Army Corps research pier are given, as is the flight plan used.

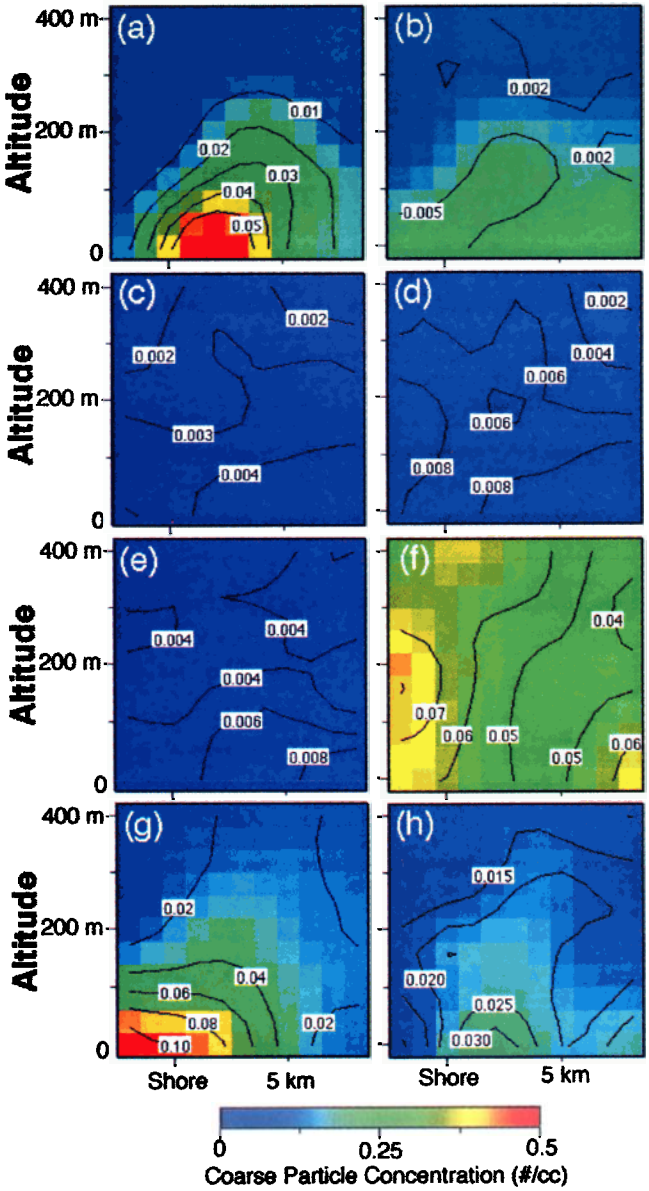

Plate 1. Near-shore 2-D cross section of particle concentration measured by the FSSP-100 in the coarse mode with diameters between 2 and 7 μm (color) and giant mode with diameters between 7 and 16 μm (isolines) for 8 flight days: (a) February 26, (b) February 27, (c) March 1, (d) March 2, (e) March 4, (f) March 8, (g) March 11, and (h) March 12. Units are in cm^{-3}.

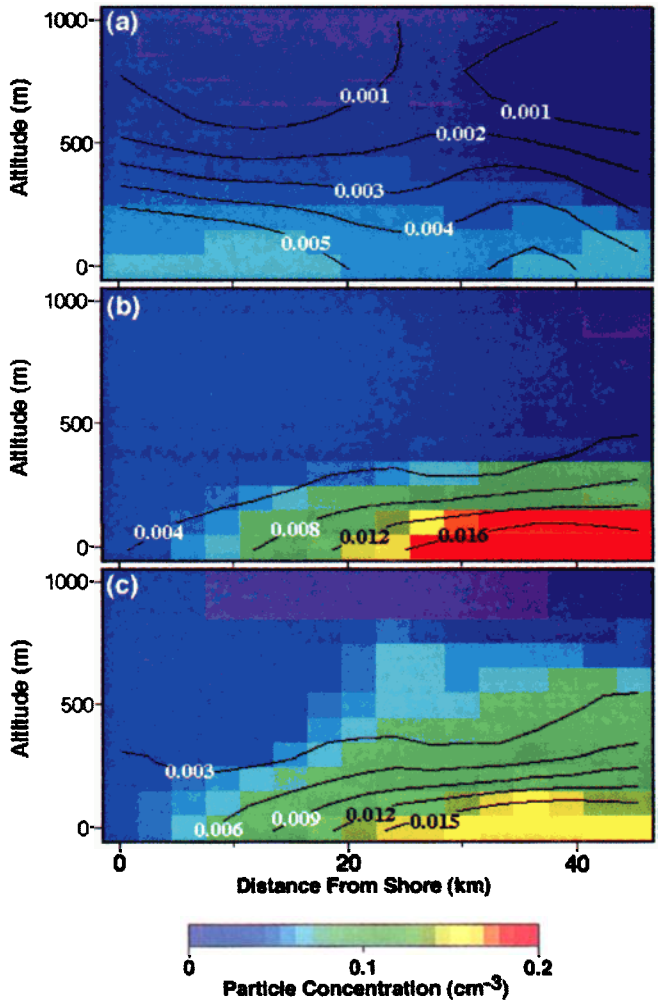

Plate 2. Nearshore 2-D cross section of particle concentration in the coarse mode measured by the FSSP-100 with diameters between 2 and 7 μm (color) and giant mode with diameters between 7 and 16 μm (isolines) for 3 days of offshore flow: (a) March 2, (b) March 1, and (c) March 4. Units are in cm^{-3}.

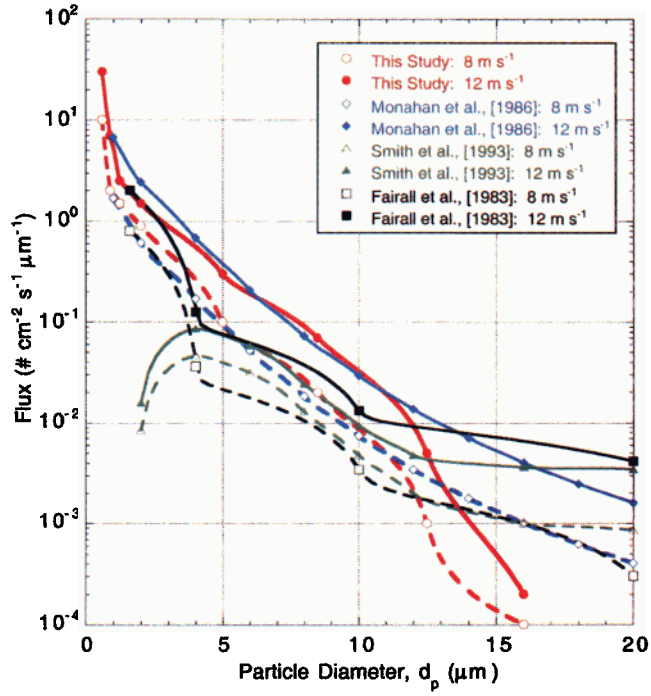

Plate 3. (opposite) Intercomparison of particle number fluxes for 8 m s^{-1} and 12 m s^{-1}. The derived values from Duck and *Fairall et al.* [1983] and the parameterizations of *Smith et al.* [1993] and *Monahan et al.* [1986] are given. Comparisons are for 80% relative humidity. Note the Monahan et al. parameterization plotted here does not include the spume term which is zero for diameter <20 μm (see text for details).

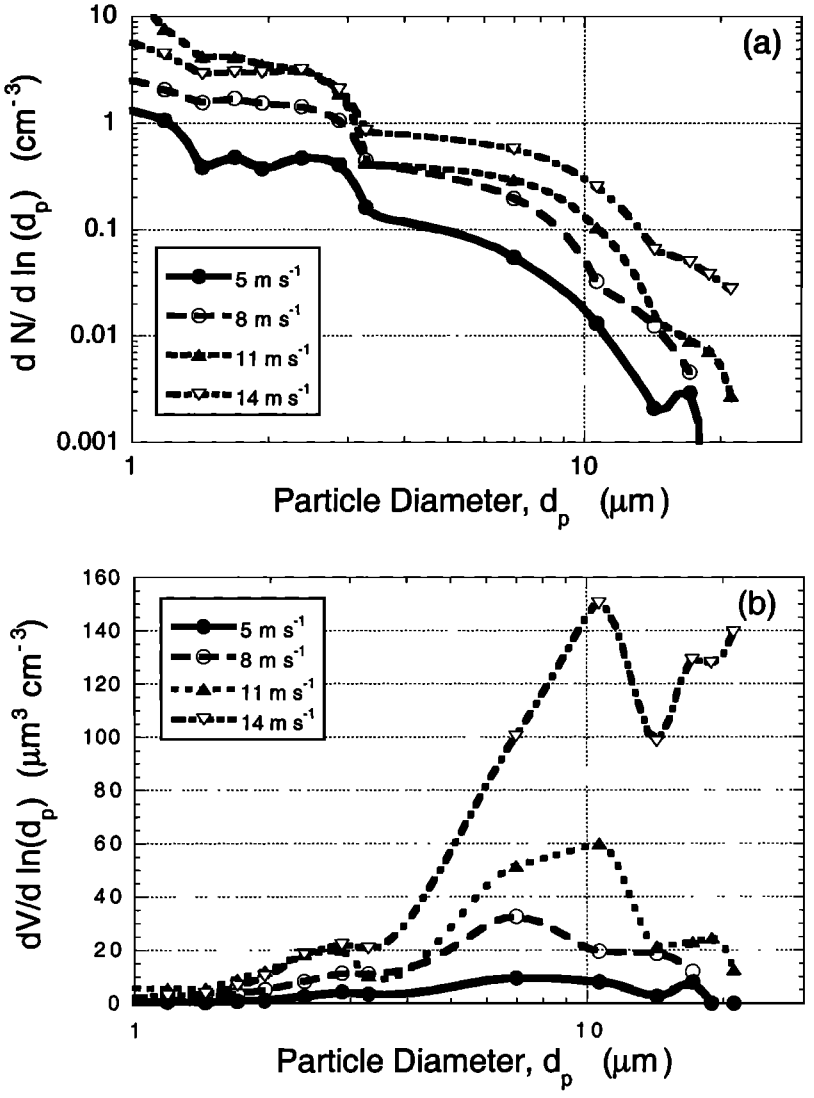

Figure 2. Thirty to one hundred meter, coarse mode sea-salt particle (a) number and (b) volume size distributions for "steady state conditions." Data from wind speeds of 5, 8, 11, and 14 m s^{-1} are given. Dried PCASP size spectra were used for diameters <2 μm.

may simply be due to the length of fetch and slight variations in the wind direction/coastline angle. The wind never went directly along the coastline. Even a relatively small 10° off the shoreline would produce a short 2 km of surf line fetch. Without a source the plume never develops vertically, and as a result, there was a short, wide plume.

3.3. Offshore Flow

The most interesting measurements during this study were obtained on days when primary sea-salt particles were not in a steady state. For March 2, 1, and 4 we experienced offshore flow at the pier with neutral stability and 10-m wind speeds of ~4, ~8, and ~12 m s^{-1}, respectively. For these cases the equivalent potential temperature was a constant to the PBL inversion height at ~1 km, indicating that the continental PBL was well mixed. Increases in particle and water vapor concentrations farther from shore were discernible, and cooling due to sensible and latent heat fluxes resulted in ~1–2°C decreases in temperature over the 40-km fetch. Relative humidity also increased by ~10–20% due to the temperature reduction and latent heat flux. While there was little change in the salt particle concentration near the shore (Plates 1c, 1d, and 1e), over much longer fetches the dynamics of this situation became much more evident.

The development of the internal marine boundary layer during these offshore flow periods is depicted in Plate 2. Plates 2a, 2b, and 2c show a two-dimensional (2-D) cross section of the coarse mode salt particle concentration (diameters >2 μm) as a function of distance from shore for the 4, 8, and 12 m s^{-1} cases, respectively. Isolines give number concentrations for salt particles >7 μm in diameter. Plate 2 shows the impact of wind speed on boundary layer development. Winds at 4 m s^{-1} produce only a few whitecaps, so particle concentrations were fairly static (Plate 2a). At wind speeds of 8 m s^{-1}, whitecaps were prevalent, and coarse particle production was clearly visible (Plate 2b). At this speed, 40 km out to sea, a low (~350 m thick) internal boundary layer had formed. This trend of increasing coarse particle production continued at wind speeds up to 12 m s^{-1} (Plate 2c), along with strong vertical transport to ~650 m, when particles larger than 7 μm were transported

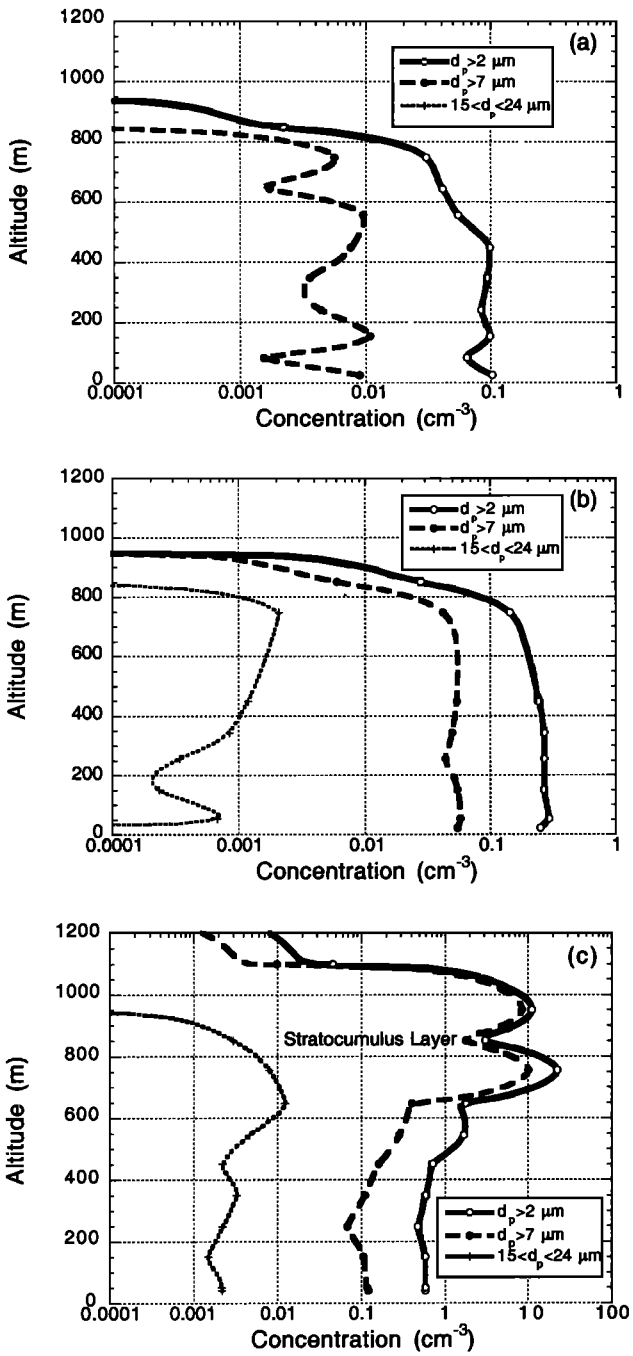

Figure 3. Vertical distribution of sea salt particles during steady state conditions for three size ranges as computed from FSSP-100 data: >2 μm, >7 μm, and between 15 and 24 μm. Three cases are given: (a) March 2, 5 m s^{-1} wind speed, (b) March 8, 11 m s^{-1} wind speed, and (c) March 11, 14 m s^{-1} wind speed.

to the top of the internal boundary layer in concentrations in excess of 1 L^{-1}.

Interestingly, in our coastal study area we found that near the surface, stronger wind speeds did not induce higher particle concentrations. While the column-integrated number of salt particles in the 12 m s^{-1} case was nearly twice that of the 8 m s^{-1} case, this increase was dominantly due to an increase in boundary layer (BL) height. The increased upward particle transport due to shear (mechanical) driven turbulence did not converge to increase particle concentrations near the surface. For the 8 m s^{-1} case the particle vertical distribution had reached steady state at ~35 km or after ~70 min of transport time (as evidenced in Plate 2b). At this point the vertical distribution of aerosols became static, implying that the upward production flux roughly equaled the downward dry deposition flux. At 12 m s^{-1} the vertical distribution of salt particles never reached such a steady state in our study area. The column-integrated particle loading was still increasing 45 km out to sea (albeit slowly). A linear projection of the 30-m particle concentration versus distance suggests that at least another ~20 km would be required before the 30-m particle concentration for the 12 m s^{-1} case equated that of the 8 m s^{-1} case (65 km total or ~90 min). An equilibrium zone would then probably be established some farther distance downwind. In either case, 1 to 2 hours of transport time was required before the marine boundary layer reached some measure of steady state.

Particle size distributions for the three offshore flow days are presented in Figure 5. The 30–100 m number and volume distributions for the shoreline (dashed) and averaged from 30–40 km offshore (solid) are given. For the 4 m s^{-1} case (Figure 5a) we see that there are only minimal changes in the size spectra. There are slight changes in particle size in the accumulation mode, and as will be shown later, they are statistically significant. We do observe some statistically significant production of coarse particles in the 3–10 μm range. This is consistent with the small number of whitecaps observed far offshore.

For the 8 m s^{-1} case (Figure 5b) we can see considerable changes in the particle size distribution as the air mass was advected off shore. A prominent volume peak is visible with a modal diameter of 7 μm. Using a least squares lognormal curve fit, we found this distribution had a volume median diameter of 6.5 μm and a geometric standard deviation (σ_{gv}) of 1.8. Sea-salt particle production is also clearly seen in the number distribution to sizes as small as 0.7 μm. As the wind speed increased to 12 m s^{-1} (Figure 5c), we also see a predominant coarse mode volume peak, although at a slightly larger size (mode of 10 μm, curve fit VMD of 8.0 μm and σ_{gv} = 1.9). We can also see fine mode sea-salt production down to 0.4 μm in diameter (note that for the fine mode here, this is for dried particles). As discussed above, in this case, there is still a net vertical transport of aerosol particles at the surface at the end of the transect.

Because of the importance of submicron salt as cloud condensation nuclei it is worth our effort to determine the minimum size threshold for salt production. Figure 6 shows the 30–100 m average particle concentrations as a function of downwind distance from the coast for our three cases. To avoid confounding the situation by the growth of aerosol particles with RH into larger size bins, data are presented for dried aerosols. Concentrations for four submicron sizes are given: 0.24 μm (0.22–0.26), 0.28 μm (0.26–0.30), 0.40 μm (0.3–0.5), 0.9 μm (0.75–1) in Figure 6a, 6b, 6c, and 6d, respectively. At the smallest size (d_p = 0.24 μm) we find that for the 4 and 8 m s^{-1} cases, surface level particle concentration actually goes down as the air mass is advected out to sea. This is in agreement with dry deposition and no/low production or, if anything, a spatial inhomogeneity of fine mode aerosol particles coming off the continent. At 12 m s^{-1}, there is an increase in particle concentration with distance, indicative of particle production, but given the magnitude of the change in concentra-

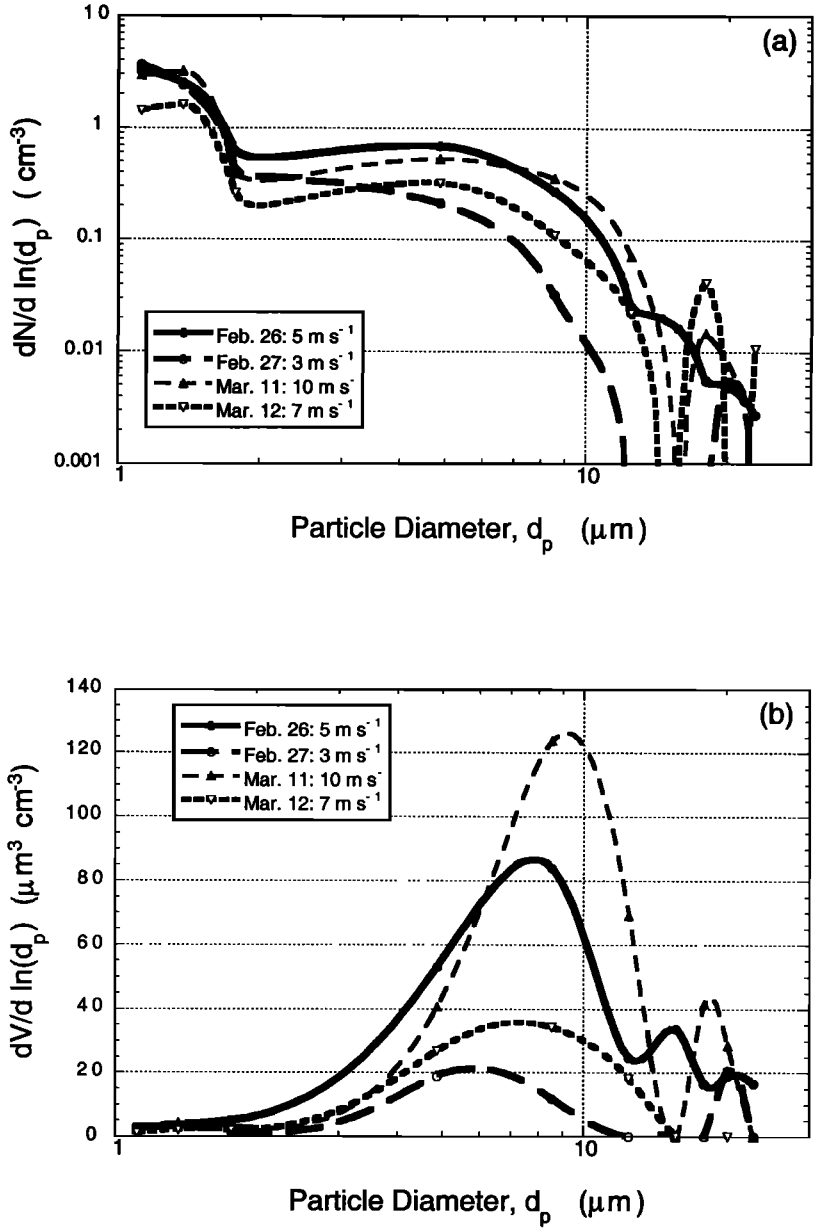

Figure 4. Size distributions for surf line generated aerosols for 30–100 m elevation: (a) number distribution and (b) volume distribution. Dried PCASP size spectra were used for diameters <2 μm.

tion (9% over 40 km) in comparison with that for the lower wind speeds, we cannot necessarily say this increase is statistically significant. However, when we go to the next larger size (d_p = 0.28 μm) at u = 4, the concentration is flat with distance, at 8 m s^{-1} the concentration is slightly decreasing with distance, and at 12 m s^{-1} the concentration is more strongly increasing with distance (13% over 40 km). Finally, at d_p = 0.9 we see increases in particle concentration for all three wind speeds. This is strongly suggestive of submicron salt particle production.

It is unlikely that any of the increase in submicron particle production in the smallest sizes (e.g., ~0.25 μm) was from gas to particle conversion. Gas phase production of sulfate from dimethyl sulfide (DMS) takes an extremely long time, of the order of a day or more, as discussed by *Quinn et al.* [1998]. Further, most of this sulfate is produced through heterogeneous cloud processing, not gas phase oxidation. Thus, for the 2 hours that we observed particles being advected offshore oxidation of DMS is not an issue. Ternary oxidation of anthropogenic SO_2 to sulfate over the ocean (with the associated high RHs) is of the order of a few percent per hour. However, these transects were performed in fairly clean conditions (aerosol optical thicknesses were below 0.07), and any particle production would almost be certainly be at sizes below the resolving limit of the PCASP. Even if we assume heterogeneous nucleation on the PCASP sized particles, it would not affect size (as volume goes as r^3).

From the trends in Figure 6 we can make a few comments about submicron sea-salt production. For wind speeds up to 8 m s^{-1}, submicron sea-salt production could not be detected for diameters <0.4 μm. (This does not necessarily mean that it does not exist, only we did not have good enough signal to

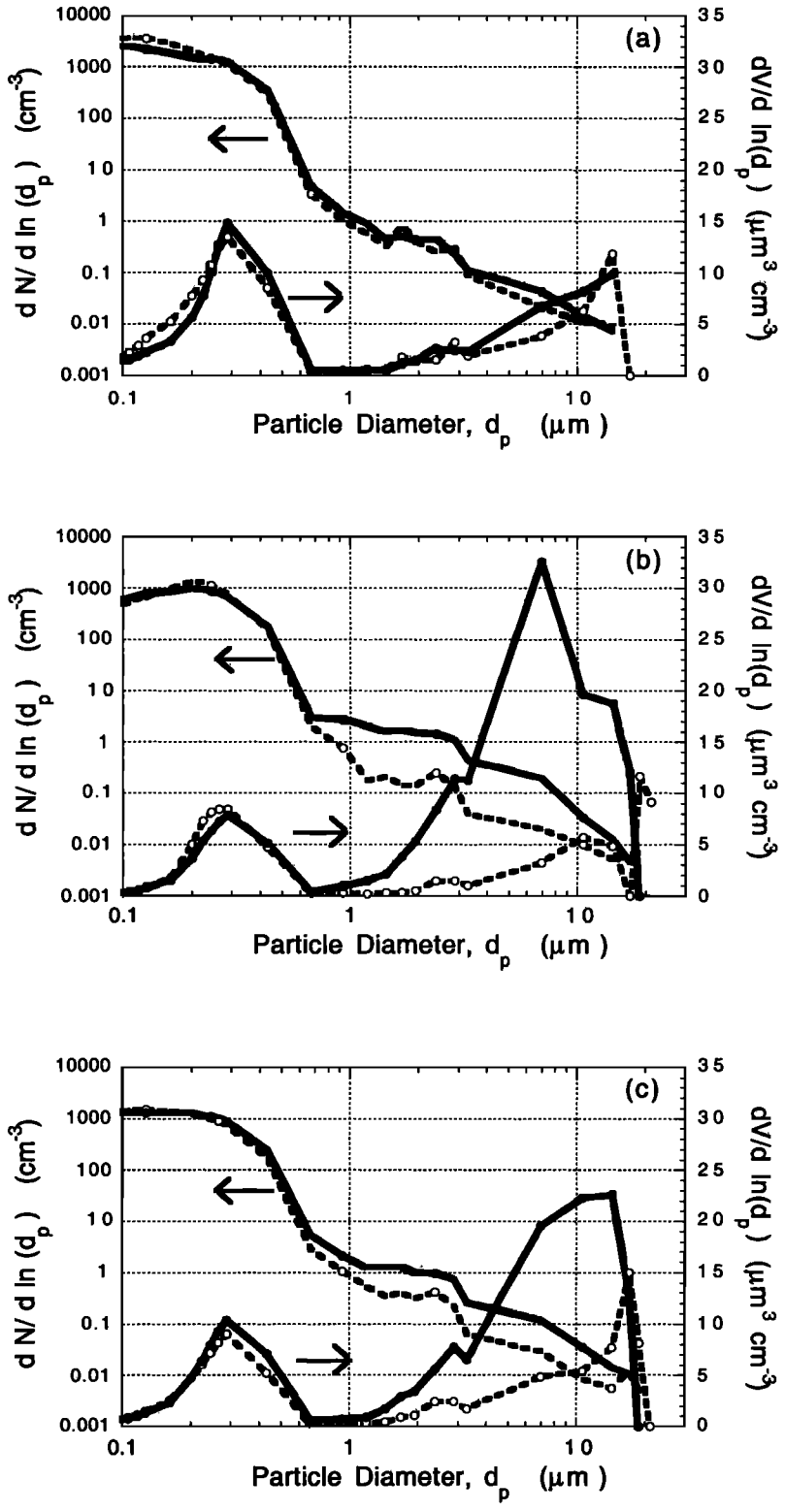

Figure 5. Thirty to one hundred meter average particle number (left axis) and volume (right axis) distributions for the studies three cases of offshore flow: (a) March 1, 4 m s^{-1} surface wind speed, (b) March 1, 8 m s^{-1} surface wind speed, and (c) March 4, 12 m s^{-1} surface wind speed. Dried PCASP size spectra were used for diameters <2 μm. The distributions at the shoreline (dashed) and 30–40 km offshore (solid) are given.

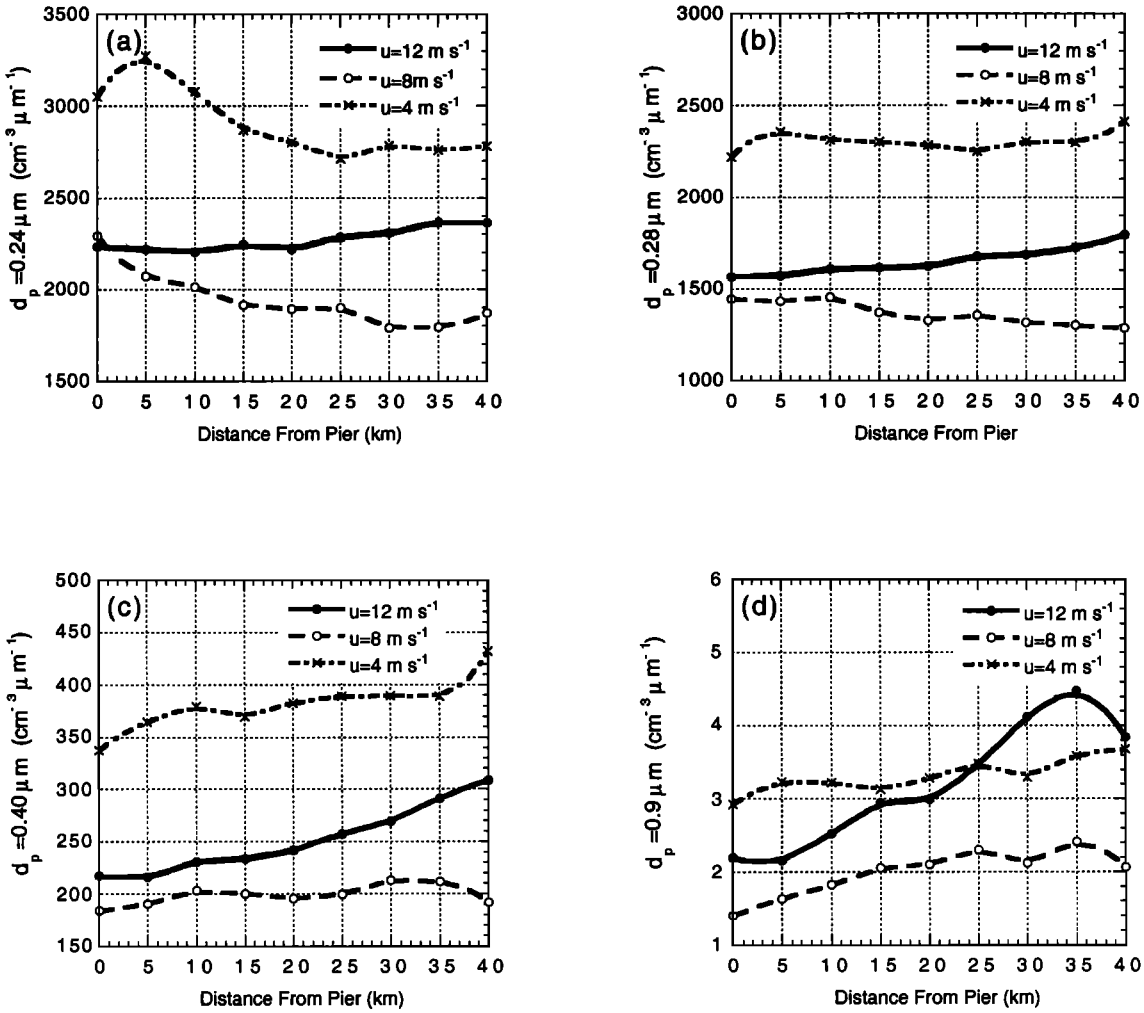

Figure 6. Thirty to one hundred meter average particle number concentration as a function of distance from the shoreline (cm^{-3} μm^{-1}). Data for four sizes (a) 0.24 μm, (b) 0.26 μm, (c) 0.40 μm, and (d) 0.88 μm are given. To avoid confounding the situation by the growth of aerosol particles with RH into larger size bins, data are presented for dried aerosols.

noise.) However, for wind speeds >12 m s^{-1}, we directly observed that primary particle production may go as low as 0.26 μm in diameter or lower. This is in agreement with *Quinn et al.* [1998], *O'Dowd and Smith* [1993], and *Murphy et al.* [1998], who over the open ocean found sea-salt particles at diameters <0.4, 0.1, and 0.1 μm, respectively.

4. Flux Estimations

The 3 days of offshore flow gave us a unique opportunity to derive salt fluxes over the ocean under "natural" conditions. By analyzing data such as presented in Plate 2 Figures 5 and 6, we can estimate the magnitude of the particle production by whitecaps. We derive the fluxes by examining surface concentration and column burden of salt particles as a function of time (or distance) downwind. In this "box"-type method the change in the column burden of salt particles (c_{col}) is simply equal to the upward flux from bubble and spume production from whitecaps (F_u) at 30 m minus the downward flux from dry deposition/sedimentation (F_d)

$$\frac{dc_{col}}{dt} = F_u - F_d = F_u - V_d c_{surf}, \quad (1)$$

where we substitute the dry deposition flux with a dry deposition velocity V_d times the surface concentration. For this calculation we use V_d values used by *Smith et al.* [1993] from *Slinn and Slinn* [1980]. Because we made our measurements in an area of active entrainment and rapid vertical transport, F_u was much greater than F_d. A sensitivity study revealed that varying V_d by a factor of 5 only resulted in a 25% change in our flux calculation. Hence this method is insensitive to the assumed value of V_d.

Table 2 presents our findings of upward flux for the 8 and 12 m s^{-1} cases (in cm^{-2} s^{-1} μm^{-1}). Particle sizes were adjusted to a relative humidity of 80%. Because particles were generated wet, hysteresis prevents much of a size change from 80% to the ambient conditions of ~55–65% for FSSP-100 data (~15% [*Gerber*, 1985]). For particles measured with the PCASP this correction was of the order of 40%. Mean values represent the mean of flux values derived for 5-km advection steps for the range of 5–35 km from shore (e.g., seven points computed at 5, 10, ..., 35 km). Errors were based on FSSP/PCASP counting uncertainties (~10%) and the signal-to-noise uncertainties for the column integration and advection. For the signal error we used the standard deviation of data points using

Table 2. Upward Sea-Salt Fluxes at an Altitude of 30 m[a]

Diameter, μm	Case	
	8 m s^{-1}	12 m s^{-1}
<0.5	NA	NA
0.6	10 ± 10	30 ± 20
0.9	2 ± 1	7 ± 2
1.25	1.5 ± 0.05	2.5 ± 0.5
2	0.9 ± 0.3	1.5 ± 0.5
5	0.1 ± 0.05	0.3 ± 0.15
8.5	0.02 ± 0.01	0.07 ± 0.03
12.5	0.001 ± 0.001	0.005 ± 0.005
16	0.0001 ± 0.0002	0.0002 ± 0.001
>18	NA	NA

[a]Units are cm^{-2} s^{-1} mm^{-1}. To make our findings comparable to other studies, we have corrected our FSSP data to a relative humidity of 80% using the *Gerber* [1985] parameterization. This produced less than 15% change in size for particles larger than 2 μm. For particles smaller, equal to, or smaller than 1.25 mm, this is about a factor of 1.4. NA indicates not available, i.e., too much noise to compute.

different interpolation schemes (linear interpolation, vertical linear interpolation, conservative kernel smoothing), plus points derived for the computed flux for each column advection step (5 km).

As can be seen in Table 2, errors were strongly size-dependent. Our highest sensitivity lies with particles with diameters in the 0.9–9 μm range, where we had high count levels and a very low background concentration. For the largest sizes ($d_p > 10$ μm), errors were predominately due to poor counting statistics, particularly for the 12 m s^{-1} case (at 12 m s^{-1} the fluxes were greater, but concentrations were lower). Also, since 30 m was the lowest our aircraft could fly, we are likely to be underestimating the flux of the largest particles, which have been shown to have a strong gradient in the lowest 30 m [*Blanchard et al.*, 1984].

For submicron particles the errors lay in the background concentration of accumulation mode aerosols. Because of these background aerosol particles we could not derive flux values for diameters <0.5 μm, although we know from Figures 5 and 6 that such production exists down to at least 0.25 μm (dry) for the 12 m s^{-1} case. However, we can say that for the 8 m s^{-1} case, there was no observable production for diameters <0.6 μm (and for this case we had very small background concentrations). Ratioing the various submicron aerosol concentrations in Figure 6 to the 0.6 μm concentration (where we do have a high signal-to-noise ratio) suggests that flux rates continue to increase for decreasing size.

5. Discussion and Hypothesis Testing

Given the limited scope of this field study, with steady state distributions at only five wind speeds and flux values for only two wind speeds under neutral stability conditions, it cannot form the basis of a parameterization scheme. Indeed, given the spread of flux estimates found in the literature, it is likely that no one parameterization or measurement set (including our own) is precise to within an order of magnitude. However, there is uniqueness to this data set which makes it valuable for adjudicating between the various existing parameterizations: (1) we directly observed the injection of sea salt into the atmosphere, (2) we operated in an area where we could determine the air mass history, and (3) we had a fairly clean continental background air mass. In sections 5.1–5.3 we discuss and compare our salt particle data with well-known parameterizations and attempt to explain observed differences. This discussion is broken down into two basic hypothesis: (1) Do our results confirm the popularly used *Smith et al.* [1993] salt flux parameterization and (2) which size distribution parameterizations or measurements presented in the literature, if any, match our results.

5.1. Hypothesis A: The Smith et al. Particle Flux Parameterization

Andreas [1998] clearly shows and discusses the high degree of variability, up to 5 orders of magnitude, in sea-salt flux values reported in the literature. The standard deviation from the mean of these 13 parameterizations is roughly a factor of 8. However, there is a group of parameterizations near the mean, with the *Smith et al.* [1993] parameterization in the middle. For particle radii between 1 and 25 μm and 14-m wind speeds up to 32 m s^{-1}, *Smith et al.* [1993] suggested the following function based on data taken in the Outer Hebrides of Scotland:

$$\frac{dF}{dr} = \sum_{i=1,2} A_i \exp\left\{ -f_i\left[\ln\left(\frac{r}{r_i}\right)\right]^2\right\}, \qquad (2a)$$

where dF/dr has units m^{-2} s^{-1} μm^{-1}, $f_1 = 3.1$, $f_2 = 3.3$, $r_1 = 2.1$ μm, and $r_2 = 9.2$ μm. A_i is given by

$$\log(A_1) = 0.0676U_{14} + 2.43 \qquad (2b)$$

$$\log(A_2) = 0.959U_{14}^{1/2} - 1.476. \qquad (2c)$$

After comparing various parameterizations, *Andreas* [1998] suggested that the Smith et al. parameterization may be low by a factor of ~3.5. Because the Smith et al. parameterization is relatively complete and the total spume droplet surface area goes as the characteristic u_*^3, *Andreas* [1998] suggested that for the 2–50 μm diameter size range the functional form be retained but with a simple 3.5 multiplicative correction factor.

Plate 3 presents our flux data for the 8 and 12 m s^{-1} cases along with findings from commonly cited studies: Smith et al. parameterization discussed above, the parameterization of *Monahan et al.* [1986] (not including the spume term which is zero for diameters <20 μm), and the measurements of *Fairall et al.* [1983]. Overall, our findings for particle fluxes are toward the higher end of values presented here but in the middle of those previously presented in the literature. Our derived particle fluxes for diameters between 4 and 10 μm are roughly similar to those of Monahan et al. and a factor of 4 higher than the Smith et al. parameterization and the measurements of Fairall et al. (similar to the factor of 3.5 suggested by *Andreas* [1998]). We are also about a factor of 10 higher than the findings of *Wu* [1992, 1993, 1994] (not shown). For larger particles ($d_p > 10$ μm) our derived particle fluxes crossover the Smith et al. and Fairall et al. For the smallest particles we derive values just slightly higher than Monahan et al.

Given the present measurements and those compared in Plate 3, it does appear that we have some confirmation of salt particle fluxes to within an order of magnitude for these limited conditions (compared to the 5 orders of magnitude shown by *Andreas* [1998]). It is also comforting that all of the methods shown in Plate 3 derive fluxes in a different way. Even so, can the these differences in measured particles be reconciled? For the most part, we feel that they can be. For particles larger than 10 μm our lower value for flux is probably due to an

Table 3. Approximate Salt Particle Size Statistics for the Film/Jet Mode Found in This Study and From the Literature for 10 m s^{-1} Winds and RH = ~80%

Reference	Location	Height, m	VMD, μm	σ_{gv}
Woodcock [1953]	subtropical Pacific	500	20	~2
Porter and Clarke [1997]	tropical Pacific	variable	11	2.3
Hoppel et al. [1989]	Tenerife	10	10	1.9–2.2
Sievering et al. [1987]	Outer Banks	variable	10	1.8–2.1
Kim et al. [1990]				
Duck (this study)	Outer Banks, North Carolina	30–100	9	1.8–2.2
Sievering et al. [1987]	Bermuda	variable	8	1.8–2.1
Kim et al. [1990]				
Smith et al. [1993]	Outer Hebrides	14	8	~2
Shettle and Fenn [1979]	composite	variable	8	2.5
Horvath et al. [1990]	U.S. East Coast	variable	7.5	2.1
Exton et al. [1986]	Outer Hebrides	10	6	~2.2
Gerber [1985]	Azores	15	6	2.0
Marks [1990]	Ireland	10	6	~2.2
Horvath et al. [1990]	Bermuda	250	5	1.7
McGovern et al. [1994]	Ireland	10	5	~2.2
Fairall et al. [1983]	southern California	10	4	~2.2
van Eijk and De Leeuw [1992]	North Sea	10	2.5	2.0
Gathman [1982]	variable	10	2	2.0

underestimation of the total coarse mode aerosol burden. These larger particles probably exist in higher concentrations below the aircraft elevation [*Blanchard et al.*, 1984]. However, the situation is more complicated for particles in the 2–10 μm diameter range. We are most confident in our flux calculations for particles in this size range as it is here that we had the best signal-to-noise ratio. Similarly, the size distributions measured by Smith et al. that went into their parameterization for this size range also had the lowest uncertainty.

We must carefully consider how and to what purpose flux measurements reported in the literature were made. Most previous studies were performed by either (1) observing the concentrations of particles in the atmosphere and estimating fluxes through dynamical constraints, such as the dry deposition velocity [e.g., *Smith et al.*, 1993; *Fairall et al.*, 1983] or (2) establishing empirical models based on whitecap coverage and dynamics coupled with bubble droplet measurements in a wind tunnel [e.g., *Monahan et al.*, 1986; *Woolf et al.*, 1987; *Wu*, 1992, 1994]. Judging the field from Figure 1 of *Andreas* [1998], neither of these methods produce systematically different results from the other. For example, *Monahan et al.* [1986] and the series of papers from *Wu* [1992, 1994]] used similar wave tank techniques but yield results toward the high and low end of reported values, respectively. In contrast, the techniques of *Fairall et al.* [1983] and *Smith et al.* [1993], both in the middle of reported values, are essentially based on the assumption that near the surface of the water the aerosol particles are in steady state and hence the upward flux is equal to the downward flux:

$$\frac{dF}{dd_p} = V_d \frac{dN}{dd_p} \tag{3}$$

where dF/dd_p is the upward flux at diameter d_p, V_d is the dry deposition velocity of the particles, and dN/dd_p is the particle number distribution.

Both wave tank and field measurement methods are based on inferences about the true atmospheric state and particle dynamics and hence are prone to uncertainty. Wave tank measurements probably cannot reproduce all surface layer effects, certainly not the 7-m swells we sometimes observed during this study. For field measurements, any error in the assumed dry deposition velocity or size distribution propagates linearly into the derived flux rates. Understanding these points, we can now explore reasons for the differences. *Andreas* [1998] gave two explanations: evaporation and gravitational settling. First, let us consider evaporation. *Andreas* [1998, p. 2179] argued that smaller particles evaporate more quickly than larger ones. "Thus, [he continues] locally-generated droplets from one size bin move to smaller size bins faster than droplets from larger size bins replenish the original bin. The small size bins are consequently underrepresented if the relative humidity is less than saturation." We reject this explanation. The evaporation of droplets is relatively fast: 30 s to 1 min at most. This explanation hinges on the idea that *Smith et al.* [1993] measured relatively young particles, which we know is not the case. *Fairall et al.* [1983] and *Smith et al.* [1991] clearly showed that the mean lifetime of large salt particle in this range was $\gg$30 min. Hence relatively rapid evaporation processes are not playing a role. Even if the sizing were off, *Smith et al.* [1993] would for the most part retrieve the correct number of particles.

Andreas' [1998] second explanation follows along the lines of air mass history as discussed above and as will be discussed in section 5.2. Simply put, larger particles come into equilibrium faster. Hence particle diameters measured for the purpose of deriving fluxes would be systematically smaller if upwind wind speeds are higher than at the measuring point. We agree this can be a problem, but we do not feel that it can be a systematic undercounting by a factor of 3.5. It would, if anything, cause an overestimate of Smith et al.'s fluxes.

Our rejection of Andreas' explanations then begs the question as to the reason of the bias. We suggest that it may mostly be due to the assumptions made when the fluxes were first

calculated. It is safe to assume that the Smith et al. size distributions are more or less sound (certainly within the factor of 3.5 asserted by Andreas). In fact, if Smith et al.'s original salt distributions were off as much as a factor of 3.5, then likely none of their conclusions would be valid and that Andreas's premis (that the Smith et al. parameterization can be a basis for another) is clearly not valid. Assuming Smith et al.'s measurements are more-or-less correct then leaves us with uncertainties in the dry deposition velocity and the question whether the measurements were made under a steady state, both of which we know to be very uncertain. Dry deposition velocities reported in literature vary by nearly an order of magnitude [*Seinfeld and Pandis*, 1998]. This is especially true for particles in the 1–5 μm in diameter range (above which V_d is simply equal to the settling velocity). Consider the complexities of a full pitching sea. Large swells and spume acting as "scrubbers" can easily increase the dry deposition velocity. Also, salt particles have varying lifetimes depending on size and precipitation and hence the air mass history must be assumed to be in steady state. As will be discussed in section 5.2, even over the open ocean this may not be a good assumption.

We must also carefully consider to what end reported measurements were made. Investigators interested in air/sea exchange and latent heat flux are interested in all droplets produced by whitecaps [e.g., *Monahan et al.*, 1986; *Wu*, 1992]. Hence, in these cases it is important to account for particles which are produced and deposited within a few seconds. In contrast, investigators interested in salt particles in the marine boundary layer are more interested in those particles which have a lifetime of several minutes to hours [e.g., *Smith et al.*, 1993; *Fairall et al.*, 1983]. The differences in these perspectives can be related to sampling height.

In conclusion of this hypothesis test, we endorse the *Andreas* [1998] suggestion that the *Smith et al.* [1993] parameterization may be low by as much as a factor of 3.5. But it is likely this factor stems not from errors in the size distribution but rather the dry deposition velocity. Hence, to be consistent, a factor of 3.5 should be included in V_d of equation (3), not dN/dr. In perspective, our own study presents data for only two cases, and it is difficult to draw general conclusions on salt particle fluxes. However, we deduce the flux of particles directly from measurements without having to infer or estimate anything about the atmospheric state variables or dynamics and carry only a slight sensitivity to uncertainties in deposition velocity. We are therefore reasonably confident in the results for these particular cases.

By considering the above points, we may begin to explain at least some of the differences in reported values of salt fluxes. First, for larger particles ($d_p > 10$ μm) our fluxes drop from being a factor of 4 higher than the Smith et al. parameterization to values that are in agreement with Smith et al. This is due to the Twin Otter flying no lower than 30 m, i.e., above the concentration maximum in the largest particles, which Smith et al. sampled. So, it would not be unreasonable if the true flux is probably a factor of 4 higher than what Smith et al. is predicting. For particles with diameters <2 μm, our values are higher simply because Smith et al. did not include them in the calculation (Smith et al. could not assume steady state for particles with lifetimes as long as these). The Smith et al. source function was limited to sizes above 2 μm and, most likely, a log-normal function should be eventually added (perhaps along the same u_*^3 line for the larger particles).

5.2. Hypothesis B: Equilibrium and the Distribution of Sea Salt

Perhaps more fundamental than the flux of coarse mode sea salt at a given wind speed is its natural size and vertical distribution in the atmosphere. It is these dependent variables which are ultimately needed to model correctly geochemical cycles, cloud dynamics, and the atmosphere's radiative balance. However, reported size distributions and concentrations in the literature vary considerably under similar conditions. Consider Table 3, where we have compiled particle size distributions reported in the literature. These values are for a nominal 10 m s^{-1} wind speed and are corrected to 80% relative humidity using the *Gerber* [1985] parameterization.

Overall, reported modal diameters of sea-salt size distributions vary by a factor of 5. Our results are in agreement with *Porter and Clarke* [1997], *Sievering et al.* [1987], *Kim et al.* [1990], *Hoppel et al.* [1989], and *Monahan et al.* [1986]. They do not agree with many others. This posses the question of which size distribution or data set, if any (including our own), should be used in marine aerosol modeling. We present three possible explanations for these differences: (1) air mass history must be explicitly and accurately accounted for before size distributions can be compared, (2) fluxes, and hence size distributions, are strongly correlated to source location and ocean water characteristics, and (3) there are large instrument uncertainties and reporting biases. Let us consider these three possibilities separately.

Collectively, published sea-salt studies suggest that one cannot simply correlate particle concentration or size to wind speed alone. For example, *Hoppel et al.* [1989] showed that in the 1–9 μm range the correlation coefficient between salt concentration and local wind speed varied between 0.4 and 0.8. Hence the regression coefficient (r^2) varied by 0.16–0.64, and thus only 16–64% of the variance in particle concentration can be explained by local wind speed alone. Comparisons in the literature of salt concentration to wind speed regressions performed by *Gong et al.* [1997] demonstrate similar variance (and note that Gong et al. only compared the mean regression lines, not the corresponding scatter used to deduce each regression line). Using a full meteorology model, Gong et al. had only slightly better results (this may simply be due to the fact that Gong et al.'s study, while using a full back trajectory model, is still fundamentally based on flux parameterizations that we know are uncertain).

5.2.1. Air mass history. Results from the literature are suggestive of a high degree of sensitivity to air mass history; how does one take into account the effect of changing environmental conditions (wind, precipitation, stability, wave characteristics etc.)? Because of the large difference in particle lifetimes in the atmosphere as a function of size, there is a possibility of biasing the size distribution of measured salt particles toward smaller sizes if wind speeds were higher upwind. This can also be demonstrated in data collected near the Outer Banks. *Kim et al.* [1990] and *Sievering et al.* [1987] observed particle VMD of ~8–12 μm in the vicinity of our own work. This is in agreement with our own findings. However, near Bermuda the VMD was only 6.5–8.5 μm for the same 10 m s^{-1} wind speed. A significant difference between these two locations is the age of the sampled air mass. Near the Outer Banks, air masses over the ocean were only a few hours old, whereas near Bermuda they were several days old. Near the shoreline, we know the air mass history, so the size distri-

bution we see is the "natural" size distribution. Any reduction in wind speed in the back trajectory will move the size distribution to smaller sizes (larger particles come into equilibrium faster than smaller particles).

This steady state/air mass history situation is much more likely to cause difficulties in midlatitudes (where most sea salt particle measurements are made) than in say the tropics. In the tropics, trade winds are for the most part steady with the occasional disruption of easterly waves. In the midlatitudes, high wind speeds are usually associated with fronts and storm systems. Thus, as air travels over open ocean through a trough or ridge to a sampling site, it is most unlikely that wind speeds (or other atmospheric parameters) would be constant for very long. In fact, the wind speed at the sampling site is unlikely to be similar to that where the salt particles are generated, especially since it is well known that storms and wind fields often intensify as they approach a coast line. Compounding this problem is the influence of precipitation scavenging and the dilution of the ocean surface by rainwater [*Marks*, 1990]. Given that *Fairall et al.* [1983] suggests that even 5-μm particles can take 24 hours to come into equilibrium with the marine boundary layer, it is likely that a form of small particle enhancement is taking place. In cases, such as ours where steady state conditions are assumed on air masses ~11 hours old, the situation may not be so grave. This is because we know that coarse mode particle concentrations are relatively low coming off the continent. There were no "background" or "legacy" salt particles to confound the measurements.

Gathman et al. [1982] attempted to compensate for air mass history by applying a term based on the 24-hour average wind speed. However, as discussed above, this is by no means physical in the midlatitudes. It is, in essence, forecasting on persistence. The another solution for the air mass history to date is that of *Porter and Clarke* [1997], which evoked dynamic modeling (*Remer and Kaufman* [1998]); that is, assigning a particle size distribution based on concentration alone (ignoring wind speed altogether). The physical explanation behind such a model is simply that concentration is implicitly related to wind speed. When wind speeds are high, equilibrium is generally reached quickly because of the dominance of very large particles. At low concentration (and hence low wind speed), particles are produced far upwind, and dry deposition or precipitation scavenging reduces the volume median diameter. While this works on climatological scales, it still poses difficulties. As discussed by *Reid et al.* [1999], dynamic modeling of aerosol particles can be dangerous as it is, in effect, a regression on a confounding variable. Such regressions are generally avoided in statistics since there is no direct physical and causal variable present. Such regressions may prove very useful (indeed Porter and Clarke's is a very useful and well reasoned parameterization), but they are by no means physically based and hence must be evoked with caution and understanding. This is particularly true when used in conjunction with models that have their own flux parameterizations (as in many general circulation models (GCMs)).

5.2.2. Organics. Next, consider a second-order possibility that particle size distributions vary naturally with sampling location over the Earth. Examination of Table 3 does not immediately bring to light any significant differences by location. Indeed, midlatitude measurements give results over the entire spectrum (although *Gong et al.* [1997] modeled systematic differences between tropical and midlatitudes due to mean meteorology differences). If any influence is present, it is most likely on smaller scales. One possibility is that coarse mode particle size is influenced by organic surfactants in the water (e.g., lipids and phospholipids). It has been suggested that organic particles are prevalent over the ocean [*Novakov and Penner*, 1993; *Rivera-Caprio et al.*, 1996; *Middlebrook et al.*, 1998; *Ellison et al.*, 1999]. These organics are correlated to sea salt, suggesting primary production. Given this fact, it is altogether reasonable to suggest that variations in the organic content of the ocean may affect the bubble bursting production of "sea-salt" particles.

5.2.3. Sampling bias. Finally, we can consider the issue of sampling uncertainty and reporting biases. It is well known that for aerodynamic reasons, coarse particles become more difficult to measure at higher wind speeds. Although this point is not an issue for open celled aircraft samplers such as the FSSP, it may become an issue for other sampling systems such as impactors or closed celled systems, for example, the active scattering aerosol spectrometer probe (ASASP). Indeed, measurements made by investigators using FSSP and FSSP derivatives provide similar results for particle size [e.g., this study; *Hoppel et al.*, 1989; *Sievering et al.*, 1987; *Kim et al.*, 1990; *Smith et al.*, 1993]. What is more troubling is that at higher wind speeds, particle size increases, making it even more difficult to measure. These biases would result in systematically smaller measured sizes. Also, one cannot ignore the impact of unintentional aerosol particle drying during sampling using closed cell systems.

There may be an unintentional biasing in the reporting of particle size by only reporting particle number or volume distributions; both the volume and number distributions should be displayed. For example, in Table 3, several of the presented values for VMD were derived from count median diameters (CMDs) and standard deviations given in the paper assuming a lognormal distributions using the Hatch-Choat equations. However, the CMDs and standard deviations were originally derived in many previous studies [e.g., *Gathman*, 1982; *Smith et al.*, 1993; *van Eijk and De Leeuw*, 1992] using a least squares fit to the number distributions. In doing so, they have a high precision in describing small particles, and low precision in describing the large particles where all of the volume (and hence mass concentration) is found. Also, because of the difficulty in sampling large particles at high wind speeds, many investigators also only sample to 2–5 μm in diameter thereby compounding the problem. Since the conversion of VMD from CMD and the standard deviation is proportional to 3 times the square of the natural log of the standard deviation for these distributions, even small errors can amplify rapidly. Hence independent determination of particle count and volume median diameters must be made.

5.3. Implications for Marine Aerosol Studies

Our study does not resolve these reporting differences in particle size. It does, however, demonstrate that some inconsistency in the literature exists for salt particle fluxes and that hypothesis B (some simple parameterization for sea-salt particles exists) fails. Perhaps this is simply an air mass history problem, and a steady state never can be assumed. No doubt, organics also play a role, although to what extent is unknown. Finally, we experimentalists must always take our measurements with "a grain of salt." It is likely that all three of these reasons and several other unknowns play a role. With these points in mind, we wish to close with two open ended questions:

1. Is there/can we develop a simple yet meaningful sea-salt

parameterization which can adequately describe sea salt and what are the key independent variables if not simply wind speed? That is, can we ever solve this through regression modeling or is a process model with more detailed air mass history and atmospheric stability required?

2. What impact do these uncertainties in reported size have on marine boundary layer/cloud modeling studies? More simply put, do these differences really matter? For example, is the *Porter and Clarke* [1997] GCM parameterization adequate for the job despite these uncertainties? Climatology, certainly? Radiation, possibly? Cloud physics, probably not?

6. Conclusions

In this paper we presented data on the evolution of the development of the marine boundary layer during marine background and offshore flow conditions. From this data set we have drawn the following conclusions:

1. At wind speeds in excess of ~6 m s^{-1} a clear internal boundary layer develops rapidly and exhibits a marked increase in coarse mode particle concentrations and both specific and relative humidity.

2. At a wind speed of ~8 m s^{-1} the sea-generated aerosol in this internal boundary layer requires at least 1 hour to come into equilibrium. At 12 m s^{-1}, nearly twice that time is probably required. Hence an area of offshore inequilibrium can exist for 60 km or more from the coast. Given the estimates from *Fairall et al.* [1983], it is likely that this zone of inequalibrium can be 2 or 3 times larger.

3. Particle fluxes presented here at 8 and 12 m s^{-1} wind speeds are derived in a more direct manner than previous studies and provide experimental validation of *Andreas'* [1998] opinion that the *Smith et al.* [1993] parameterization may be low by as much as a factor of 3.5. This correction in the *Smith et al.* [1993] parameterization is most likely due to an underestimation of the dry deposition velocity and not other factors such as the measured size distribution. Hence in order to correctly obtain the correct concentration of salt particles found by Smith et al., one must be very careful when applying this correction and make sure both the upward and downward fluxes are scaled appropriately.

4. There are large differences in the literature for sea-salt size distributions, and note that only half of the variance in salt particle concentration can be explained by local wind speed alone. These differences are likely due to three reasons: (1) natural variability in the ocean's wind and precipitation fields prohibit smaller sea-salt aerosols from ever getting into steady state, (2) variations in the insoluble organic content of the ocean influences the bubble bursting process and hence the salt particle size distribution, and (3) instrumentation/presentation biases.

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