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# Reconciliation of coarse mode sea-salt aerosol particle size measurements and parameterizations at a subtropical ocean receptor site

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[1] In August/September of 2001, the R/P FLIP and CIRPAS Twin Otter research aircraft were deployed to the eastern coast of Oahu, Hawaii, as part of the Rough Evaporation Duct (RED) experiment. Goals included the study of the air/sea exchange, turbulence, and sea-salt aerosol particle characteristics at the subtropical marine Pacific site. Here we examine coarse mode particle size distributions. Similar to what has been shown for airborne dust, optical particle counters such as the Forward Scattering Spectrometer Probe (FSSP), Classical Scattering Aerosol Spectrometer Probe (CSASP) and the Cloud Aerosol Spectrometer (CAS) within the Cloud Aerosol and Precipitation Spectrometer (CAPS) instrument systematically overestimate particle size, and consequently volume, for sea salt particles. Ground-based aerodynamic particle sizers (APS) and AERONET inversions yield much more reasonable results. A wing pod mounted APS gave mixed results and may not be appropriate for marine boundary layer studies. Relating our findings to previous studies does much to explain the bulk of the differences in the literature and leads us to conclude that the largest uncertainty facing flux and airborne cloud/aerosol interaction studies is likely due to the instrumentation itself. To our knowledge, there does not exist an in situ aircraft system that adequately measures the ambient volume distribution of coarse mode sea salt particles. Most empirically based sea salt flux parameterizations can trace their heritage to a clearly biased measurement technique. The current “state of the art” in this field prevents any true form of clear sky radiative “closure” for clean marine environments.

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## 1. Introduction

[2] It has been repeatedly shown that measurements of wind speed–dependent sea-salt concentrations and sea spray fluxes reported in the literature vary by several orders of magnitude [e.g., *Andreas*, 1998; *Gong et al.*, 1997; *Lewis and Schwartz*, 2004]. Figures in the work of *Porter and*

*Clarke* [1997], *Reid et al.* [2001, Table 3] and *Smirnov et al.* [2003, Figure 4] listed many studies and found that reported volume median diameters (VMDs) varied by over a factor of 5. Even the application of fundamental processes such as sea-salt production and dry deposition have come increasingly into question [e.g., *Reid et al.*, 2001; *Hoppel et al.*, 2002]. The uncertainty is compounded with the community's realization that the wind–whitecap relationship is highly variable with such additional independent variables as wind/wave direction, sea surface temperature and chemistry [*Terrill et al.*, 2001; *Mårtensson et al.*, 2003].

[3] It is unclear to what extent fundamental measurements of sea-salt fluxes in wave tanks, inferred fluxes from receptor modeling, or even dry deposition estimates are valid. Given that sea-salt size is likely dependent on a tremendous number of natural variables (e.g., age, RH, bubble dynamics, possible surfactants in the droplets) and given the added complexity of measuring sea salt in the marine environment, some divergence in reported size should be expected. However, systematic difference factors of two or more are outside the realm of reasonable results. During the PRIDE field study, *Reid et al.* [2003] found

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**Table 1.** List of Coarse Mode Sea-Salt Volume Distributions From the Literature<sup>a</sup>

Reference	Location	RH	Height, m	VMD, $\mu\text{m}$	$\sigma_{\text{gv}}$	Explained?
<i>Aerodynamic Particle Sizers</i>						
Clarke <i>et al.</i> [2003]	Hawaii	75%	14	$\sim 5$	$\sim 1.7$	yes
Maring <i>et al.</i> [2003a]	Puerto Rico	dry	10	4/5	2	yes
Quinn <i>et al.</i> [1996]	southwestern Pacific	55%	10	3/4	1.8	yes
This study	Hawaii	dry	15	2.9/4	1.7	yes
<i>Cascade Impactors</i>						
Hoppel <i>et al.</i> [1989]	Tenerife	Amb	10	9	1.9–2.2	yes
Howell and Huebert [1998]	ASTEX/Atlantic	Amb	cliff	7	$\sim 1.9$	no
Marks [1990]	Ireland	Amb	10	4.5	$\sim 2.2$	yes
McGovern <i>et al.</i> [1994]	Ireland	Amb	10	5	$\sim 2.2$	yes
Quinn <i>et al.</i> [1996]	SE Pacific	55%	10	2.7/4	1.82	yes
Quinn <i>et al.</i> [2000]	Composite ACE-1&2	55%	surface	2.5/4	2	yes
Reid <i>et al.</i> [2003]	Puerto Rico	Amb	$\sim 10$	$\sim 4$	2	yes
D. Savoie (unpublished data, 2000) <sup>b</sup>	Puerto Rico	Amb	10	4	2	yes
<i>Optical Particle Counters</i>						
Collins <i>et al.</i> [2000]	ACE-2/Tenerife	Amb	variable	$\sim 8$		yes
Exton <i>et al.</i> [1986]	outer Hebrides	Amb	10	6	$\sim 2.2$	yes
Gathman [1982, 1983] <sup>c</sup>	variable	Amb	10	2.1	2.0	insuf. info.
Gras and Ayers [1983]	Cape Grim		10	2	$\sim 2$	
Fairall <i>et al.</i> [1983] and Schacher <i>et al.</i> [1981]	JASIN	Amb.	10	4	$\sim 2.2$	yes
Gerber [1985]	Azores	Amb	15	6	2.0	yes
Horvath <i>et al.</i> [1990]	Bermuda	Amb	250	5	1.7	insuf. info.
Horvath <i>et al.</i> [1990]	U.S. east coast	Amb	variable	7.5	2.1	yes
Kim <i>et al.</i> [1995]	ASTEX	Dry	10	1/2	1.5	
Reid <i>et al.</i> [2001]	Outer Banks, NC	Amb	30–100	10	1.8–2.2	yes
Sievering <i>et al.</i> [1987] Kim <i>et al.</i> [1990]	Outer Banks, NC	Amb	variable	8	2.1	yes
Shettle and Fenn [1979]	composite	Amb	variable	8	2.5	yes
Sievering <i>et al.</i> [1987] and Kim <i>et al.</i> [1990]	Bermuda	Amb	variable	5.6	1.7	insuf. info.
Smith <i>et al.</i> [1993]	outer Hebrides	Amb	14	8	$\sim 2$	yes
van Eijk and de Leeuw [1992] <sup>d</sup>	North Sea	Amb	10	2	2.0	yes
van Eijk and de Leeuw [1992] <sup>e</sup>	North Sea	Amb	10	8	2.0	yes
This study	Hawaii	Amb	variable	8	1.5	yes
<i>Inversions (Ambient)</i>						
Smirnov <i>et al.</i> [2003]	Midway, Lanai, Tahiti	Amb	integrated	6	2	yes
This study	Lanai	Amb	integrated	6	2	yes
<i>Other</i>						
Woodcock [1953] and Porter and Clarke [1997]	subtropical Pacific	Amb	500	18	$\sim 2$	yes

<sup>a</sup>For sized distributions listed for anything other than ambient conditions, the VMD is given for the measurement/80% RH.<sup>b</sup>Unpublished data by Savoie from the PRIDE campaign but discussed in the work of Reid *et al.* [2003].<sup>c</sup>Gathman also gives two modes. The larger mode at  $\sim 20 \mu\text{m}$  diameter is physically related to “spume.”<sup>d</sup>Only the number distribution is given in the manuscript and the listed VMD is approximated from the Hatch Choat equations. The column “Explained” reports if the finding is explainable based on the findings of this study. “Insuf. info” indicates that there is insufficient information in the cited manuscript to make this determination.<sup>e</sup>On the basis of re-examination of raw volume data.

widely varying reports of coarse mode dust particle size and traced such differences to specific systematic instrumentation biases. They also found that significant potential exists for systematic error in sea-salt particle measurement. However, in the analysis of Reid *et al.* [2001], a clear delineation for sea salt based on measurement type was not cleanly detected. This result is updated in Tables 1 and 2 of this manuscript, with measurements separated by type. Clearly, there is no clean separation between measurements type, with VMD estimates ranging from 3–8  $\mu\text{m}$  for almost every method. Can this variability be reconciled?

[4] As part of the Rough Evaporation Duct (RED) experiment, the R/P Floating Instrument Platform (FLIP) and the Center for Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter research aircraft were deployed to the eastern (windward) shore of Oahu, HI to study air-sea fluxes of the subtropical marine boundary layer [Anderson

*et al.*, 2004]. During RED we wished to examine whether currently used source/sink functions and size parameterizations can adequately explain sea-salt particle concentrations at a subtropical receptor site. The principal goal of this manuscript is to assess how coarse mode sea-salt size distributions vary in a clean marine site and utilize our findings to explain the apparent variability in the literature. This will principally be done through comparisons of all particle-sizing instruments during a series of Twin Otter flybys of FLIP. Here we consider particle size distributions in 0.6 to 15  $\mu\text{m}$  diameter range derived from the surface and airborne aerodynamic particle sizers, FSSP/CSAPs, and PCAPs, with the addition of the CAPS/CAS probe data from the CIRPAS Twin Otter. We also examine retrievals from AERONET Sun photometer sites in the region using the Dubovik and King [2000] and O'Neill *et al.* [2001] algorithms. This range is considered to be inclusive of the “coarse mode” and includes the bulk of

**Table 2.** List of Instruments Used in This Study<sup>a</sup>

Instrument	Model	Type	Diameter Range, $\mu\text{m}$	Sampling
<i>R/P FLIP</i>				
Aerodynamic Particle Sizer (APS)	APS 3320	AS	0.6–15	dry
CSASP	PMS HV-100	OPC	0.5–40	ambient
Filter Samples	—	Grav/Chem	<12	dry
FSSP-100	DMT SP-100	OPC	1–32	ambient
PCASP	DMT SP-200	OPC	0.1–3	dry
<i>CIRPAS Twin Otter</i>				
Airborne Aerodynamic Particle Sizer (AAPS)	APS 3320 derivative	AS	0.6–<8	ambient
CAPS	DMT CAS	OPC	0.4–62	ambient
FSSP-100	DMT SP-100	OPC	1.5–40	ambient
PCASP	DMT SP-200	OPC	0.1–3	dry
<i>AERONET</i>				
Sun photometer Coconut Island and Lanai	<i>Dubovik and King [2000]</i>	inversion	0.1–15	column-integrated
Sun photometer Coconut Island and Lanai	<i>O'Neill et al. [2001]</i>	spectral deconvolution	Fine/Coarse AOT	column-integrated

<sup>a</sup>Sampling refers to whether the aerosol particle size distribution was performed under assumed “ambient” conditions, or whether an inlet heater was present to dry the sample humidity to <35%. AERONET Sun photometer inversions yield column-integrated values.

the sea-salt mass from the bubble busting/film drop and jet drop mechanism. It is these particles that transport the vast majority of sea-salt mass across the world's oceans. We do not discuss giant spume droplet mode, even though its tail sometimes falls into this category. We go to great lengths to clearly record found biases, and corroborate the many anecdotal stories of aerosol sampling performance that are often discussed but rarely documented. We end with a discussion of the systematic biases detected in the literature and how errors propagate through measurement and model systems.

## 2. Mission Summary, Methods, and Approach

[5] Conducted in late August to early September 2001, the Rough Evaporation Duct (RED) experiment was an Office of Naval Research Code 322 sponsored field campaign to study the air-ocean boundary over the subtropical ocean. A complete description of the mission including photographs can be found in the work of *Anderson et al. [2004]* and only a brief summary is presented here. A summary of methods evaluated here is presented in Table 2.

### 2.1. R/P FLIP

[6] Central to the RED mission was the deployment of the Scripps Institution of Oceanography Marine Physical Laboratory's Research Platform Floating Instrument Platform (R/P *FLIP*), which was moored 11 km off the northeast (windward) shore of Oahu, Hawaii from 21 August to 14 September 2001 (21.6836° N, 157.8357° W). The R/P *FLIP* is a 114 m research platform which, when deployed, has its stern flooded, thus turning it on end and creating what is effectively a manned spar buoy. While designated nonpropelled, *FLIP* does have thrusters to adjust direction. If uncorrected, *FLIP*'s back naturally turns into the wind.

[7] Aerosol and rapid response state variable data discussed in this paper were collected on the starboard boom. Located 11 m above the ocean surface, and 8 m off the side of the *FLIP*, instrumentation was deployed via a mobile trolley and could be retracted for periodic calibration and maintenance. The trolley included a Forward Scattering Spectrometer Probe (FSSP-100) measuring particle sizes

between 1 and 32  $\mu\text{m}$  diameter, and a Passive Cavity Spectrometer Probe (PCASP-100X) measuring particle sizes between 0.1 and 3  $\mu\text{m}$  in diameter.

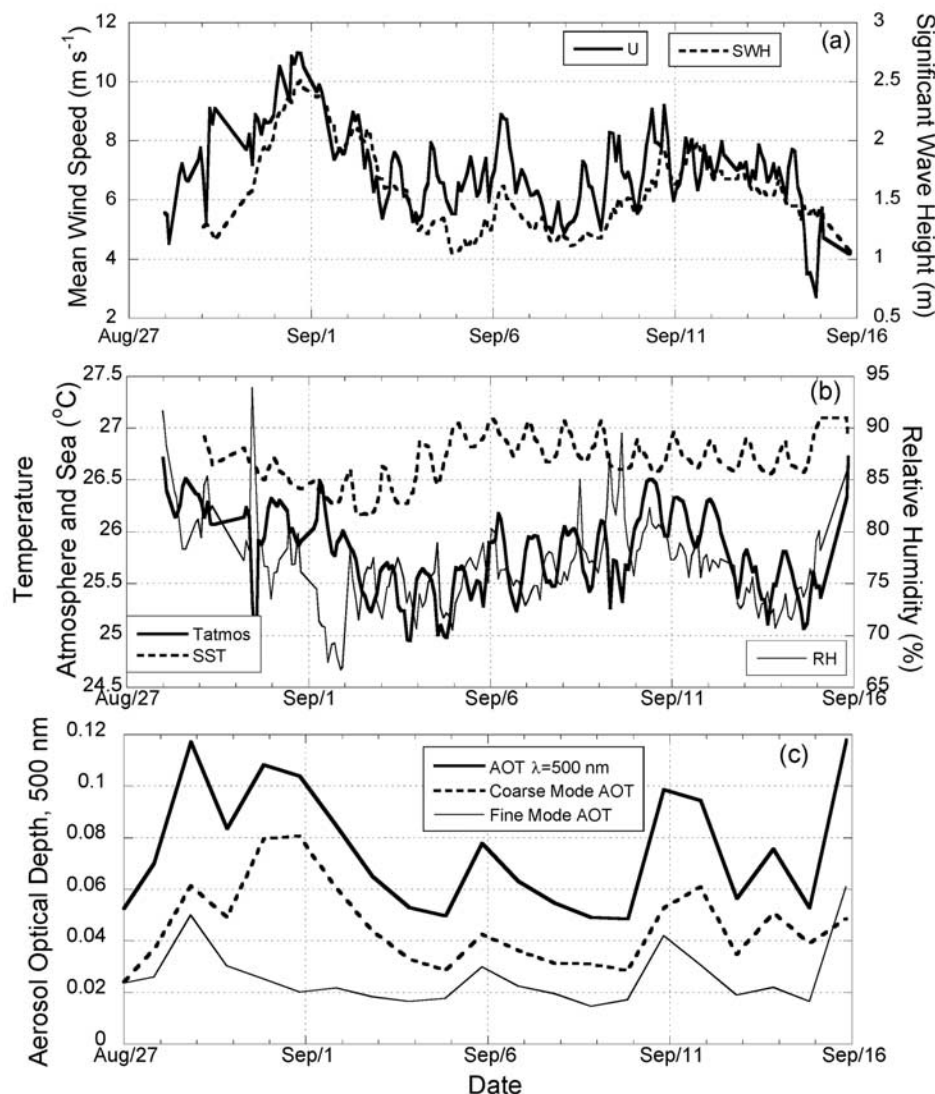
[8] The PCASP-100X and FSSP-100 were pointed into the wind and ventilated using auxiliary pumps. During the RED study, *FLIP*'S back was generally aligned to 270 degrees, the predominant wind direction of the persistent subtropical trade winds. From this orientation, *FLIP* was allowed to rotate freely such that its back always faced into the wind. Consequently, *FLIP*'s port and starboard booms projected from the main superstructure nearly normal to the prevailing wind (because the port boom is larger, there was a slight 10 degree offset). As the trade winds of the region were mostly steady out of the east, at no time was the difference between wind direction and the particle inlets in excess of 20 degrees. This is well within the 30-degree limit found in previous analyses by M. Smith (University of Leeds, personal communication, 1998).

[9] Both the FSSP-100 and PCASP-100x underwent the Droplet Measurement Technologies, Inc. electronics upgrades (SPP-100 and SPP-200, respectively) prior to deployment. Detailed descriptions on the theory and operation of these instruments can be found in the work of *Reid et al. [2003]*. The inlet-deicing heater for the PCASP was not used on *FLIP* during the study, but the instrument still produces a nearly dry particle size distribution than ambient (at least 30% RH depression; *Strapp et al. [1992]* and discussed later).

[10] For completeness, we occasionally use data from the TNO Classical Scattering Aerosol Spectrometer Probe, the lab bench version of the FSSP mounted on a center mast for additional comparisons. CSASP is similar to the FSSP but with original PMS electronics, has its own blower, and provides data in all four gain stages simultaneously. That is, it gives 4 separate size spectrums from each of its four amplifiers (3.5–48, 3–32, 1.5–15, and 0.5–8  $\mu\text{m}$  diameter for gains 0–3, respectively). Consequently, a curve fit must be applied to the data. This instrument was placed on a mast 8 m above the NRL FSSP with a sampling horn pointed into the wind. The CSASP is discussed in detail by *de Leeuw et al. [2003a]*.

[11] Also used in this manuscript were a TSI Aerodynamic Particle Sizer 3320 (APS,  $0.7 < d_{ac} < 20 \mu\text{m}$ ) and a





**Figure 1.** Time series of aerosol and meteorology variables onboard the R/P *FLIP*. (a) Mean wind speed and significant wave height. (b) Atmospheric and sea surface temperature and atmospheric relative humidity. (c) Daily averaged aerosol optical depth (AOT) from the AERONET site at Coconut Island, HI.

filter sampler. For the APS, air was provided by a 5" inlet pipe that protruded 2 m above the top of *FLIP* (16 m above the ocean) and operated at a mean flow of 400 lpm. Sample air was heated to reduce relative humidity to below 30% before feeding the APS. APS data were corrected based on the findings of *Armendariz and Leith* [2002] for summing mode.

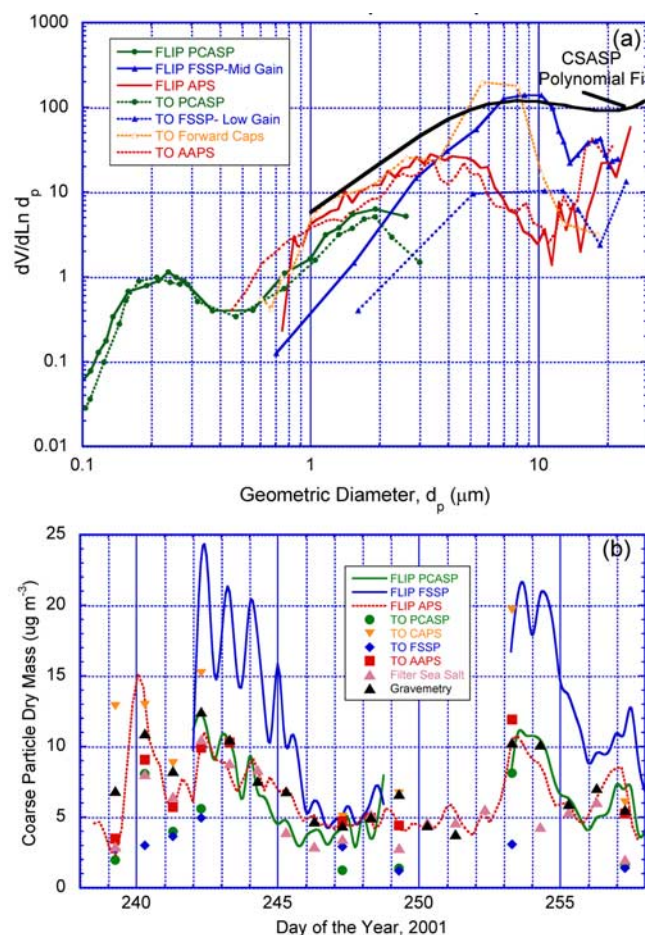
[12] Teflon filter samples were taken for 4-hour periods each day (during the time period of Twin Otter flights) and subjected to gravimetric analysis, ion chromatography, inductively coupled plasma atomic emission spectrography, and electrospray ionization–ion trap mass spectrometry [*Crahan et al.*, 2004]. Filters fed off of a separate knife-edge inlet facing into the wind with a face velocity of  $9\text{ m s}^{-1}$ .

## 2.2. Aircraft Operations

[13] The CIRPAS Twin Otter carried a basic navigation and meteorological package. Particle instruments on board

included FSSP-100 and PCASP-100X probes similar to those deployed on *FLIP* (although the TO FSSP was set on a lower gain setting such that particle size was measured between 1.5 and  $52\text{ }\mu\text{m}$ ). General descriptions, data and results from the humidigraph can be found in the work of *Crahan et al.* [2004].

[14] Also on the Twin Otter wing pods were the DMT Cloud Aerosol and Precipitation Spectrometer (CAPS), and airborne Aerodynamic Particle Sizer (AAPS) probes. Sea salt particle size distributions were measured with the Cloud Aerosol Spectrometer (CAS) portion of the CAPS probe. CAS is an open celled instrument and nominally infers particle size between 0.4 and  $62\text{ }\mu\text{m}$  in diameter in 20 channels by measuring particle scattering between  $5^{\circ}$  and  $14^{\circ}$  in both forward (herein FCAPS) and backscatter (BCAPS) detectors (individual size distributions are given for forward and back scattering separately). Early results from this study show that the BCAPS is totally unsuited for aerosol work and thus it is not discussed further in this manuscript.



**Figure 2.** (a) Particle size intercomparison from CIRPAS and *FLIP* instruments for the 10 September flyby. (b) Time series of inferred coarse mode particle dry mass from the instruments used in this study. Continuous reading instruments were subjected to 6-hour averaging. CSASP data (not shown) track the *FLIP* FSSP almost perfectly although the amplitude is fairly subjective.

[15] The wing mounted AAPS derives its basic design from the APS 3320 similar to the one on the R/P *FLIP*, but does have important differences in internal plumbing. Specifically, the pump line is significantly longer than the bench-top version. A complete description of this instrument can be found in the work of Wang *et al.* [2002].

### 2.3. Other Data

[16] The AERONET Sun photometers measured spectral aerosol optical thickness (AOT,  $\tau_a$ ) at 7 wavelengths (340, 380, 440, 500, 675, 840 and 1020 nm) plus column-integrated water vapor from a 960 nm channel [Holben *et al.*, 2001]. Cloud screening was performed using the algorithm of Smirnov *et al.* [2000]. We used the Dubovik and King [2000] algorithm to assess column-integrated size distribution, and the algorithm of O'Neill *et al.* [2001] to separate out fine versus coarse mode optical depth. Back trajectories were performed using the NOAA Real-time Environmental Applications System Hybrid Single-Particle

Lagrangian Integrated Trajectory (see <http://www.arl.noaa.gov/ready/hysplit4.html>).

### 3. Intercomparison of Sizing Methods

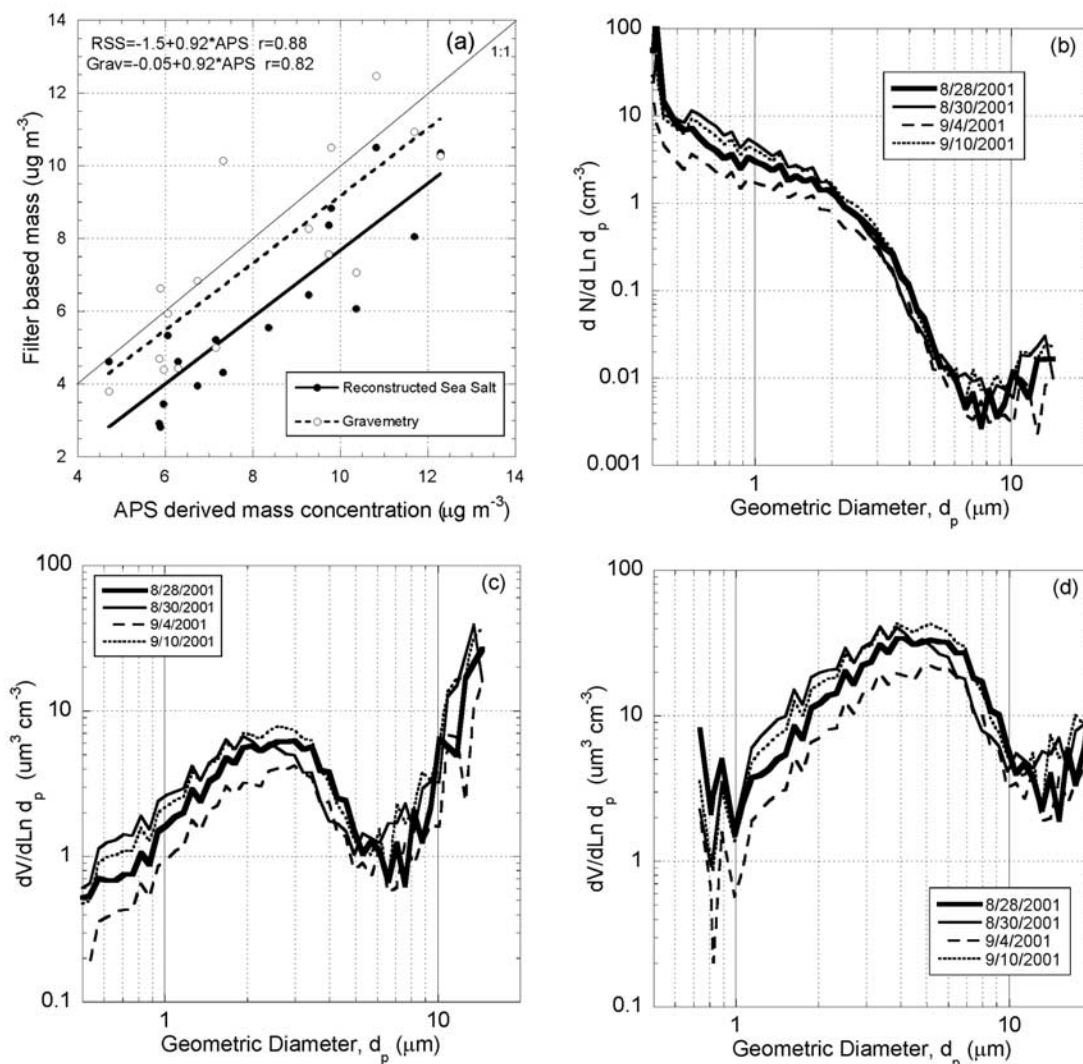
[17] A complete description of the atmospheric conditions and fluxes will be in a forthcoming paper, and some further information can be found in the work of Anderson *et al.* [2004]. In summary, data were collected on *FLIP* from the period of 28 August to 14 September 2001 and on nine occasions the Twin Otter performed vertical profiles over the vessel. Figure 1 presents meteograms for the *FLIP* as well as an AERONET time series from the Coconut Island site, 10 km away. During the mission, the atmosphere can be considered “background” subtropical marine conditions with scattered cumulus, winds varying between 2 to 12  $\text{m s}^{-1}$ , slightly unstable conditions, marine boundary layer heights of  $\sim 500$  m and ambient relative humidity in the 70–85% range. AERONET optical depths are in the realm of what are considered “clean marine background” by Smirnov *et al.* [2003]. On 28 August, we do suspect that the atmosphere was impacted by the Kilauea volcano based on HYSPLIT trajectories, filter chemistry of Crahan *et al.* [2004] and anomalous fine/coarse mode fractionation.

#### 3.1. cursory Comparisons of Observations of Particle Size and Volume

[18] In our comparisons we have to proceed from two primary postulates. The first is that the filter data from Crahan *et al.* [2004] are correct. This or any other primary standard in a field trial must be taken with a certain degree of faith. Second, we must assume some initial form of the hygroscopic growth curve. By necessity we have to compare measurements of dry and ambient salt particles. For this study, we use the empirical parameterization of Gerber [1985]. Forms similar to these make up the bulk of citations in the literature for this purpose, and vary  $d_{\text{amb}}/d_{\text{dry}}$  by  $\pm 10\%$ . It is recognized that recent studies, including Crahan *et al.* [2004], suggest that even small additions of organics can suppress hygroscopic growth in ambient sea-salt particles. Given the highly nonlinear response at high RH values, this could be an issue. However, as is also discussed later in the manuscript, this suppression actually makes comparisons diverge further. By using the Gerber parameterization, we will have errors toward more hygroscopic growth than is probably there and hence are in essence giving instruments “the benefit of the doubt” when computing particle dry sizes. In addition to these two postulates, there are a number of assumptions that must be made about particle microphysical properties, including density, index of refraction, and chemistry that we take from Tang *et al.* [1997].

[19] To initiate our comparison, consider the sea-salt event of 10 September. Back trajectories and models suggest this event was due to a combination of slight local production coupled with significant production greater than 72 hours up wind [Caffery *et al.*, 2004]. This event brought sea salt to the region on a large scale, and is likely the best day for comparisons for all methods. In Figure 2a we present volume distributions for the APS, PCASP and midgain FSSP on the *FLIP*, and the AAPS, PCASP, and forward CAPS (FCAPS) probe on the aircraft averaged over





**Figure 3.** Evaluation of the APS particle number and volume distributions from the *FLIP* APSs. (a) Regression of APS-derived mass to filter-based mass from gravimetry and reconstructed sea salt. (b) Number distribution (dry), (c) volume distribution (dry), and (d) inferred ambient volume distribution.

a 2-minute period. Also shown in the dark solid line is the polynomial curve fit of the 4 gain spectrums of the TNO CSASP. For this plot, all coarse mode particle size distributions were adjusted to ambient relative humidity ( $\sim 76\%$ ) and an assumed refractive index of 1.36. PCASP measurements are at their native relative humidity (as will be discussed, almost certainly dry).

[20] Large differences are visible within Figure 2a. While fine mode particle measurements from the PCASPs track each other extremely well, the optical particle counters and aerodynamic particle sizers for the coarse mode show strong systematic divergence. The APS systems give values of volume median diameter (dry) on the order of  $2.5\text{--}3\ \mu\text{m}$ , corresponding to a VMD of  $\sim 4\text{--}5\ \mu\text{m}$  as shown in the figure. Conversely, the midgain FSSP, CSASP fit, and the FCAPS probe give much larger values for VMD ( $\sim 7\text{--}8\ \mu\text{m}$ ) and consequently have much larger total volumes.

[21] The differences found on 10 September were endemic for the entire study period. Figure 2b presents estimated coarse mode mass (dry) concentrations for the entire study.

Each of the methods track very well, although significant amplitude shifts exist, and like instruments behave similarly. In the following subsections, we examine each class of instrument and analyze their performance in the context of Figure 2. In the interest of brevity, complete descriptions of particle measuring instruments, their calibration, and typical first-order corrections can be found in the work of Reid *et al.* [2003]. We note differences where appropriate.

### 3.2. *FLIP* APS

[22] Aerodynamic particle sizers (APS) assess a particle's size by its ratio of particle drag to mass, or essentially, the time it takes for a particle to adjust its velocity to its surrounding flow. APS systems currently on the market are based on the measured velocity of a particle flowing through a jet; faster particles achieved the flow speed more rapidly and hence are aerodynamically smaller.

[23] APS systems have several issues worth discussing briefly. First, calibration studies have suggested that the APS under-sizes particles with irregular shapes, such as dust

[e.g., Marshall *et al.*, 1991]. A very good synthesis of these issues is presented in the work of Wang *et al.* [2002]. For particles such as dry sea salt, under-sizing may be as large as 10%.

[24] The APS 3320 used in this study varies significantly from the 3310 used in the work of Reid *et al.* [2003]. Most notably, Armendariz and Leith [2002] found significant errors in counting efficiencies. These varied to as much as a factor of 40% in the region where the volume median diameter of dry sea salt exists ( $2 < d_{ac} < 4 \mu\text{m}$ ). We corrected our data using their parameterizations for summing mode. The APS 3320 also has a significant positive artifact for particles greater than  $\sim 10 \mu\text{m}$  in aerodynamic diameter due to the recirculation of particles through the sheath flow back into the viewing volume [Stein *et al.*, 2002; D. Covert, University of Washington, personal communication, 2000]. This issue is acknowledged by TSI Inc., and has been corrected in the latest version of these instruments (APS 3321). Because the APS inlet system has a cut point  $\sim 12 \mu\text{m}$ , the lack of particles greater than this size (say from spume production), would not manifest itself in this instrument. Regardless, for our study winds were  $< 12 \text{ m s}^{-1}$  and spume production was minimal during 90% of the study.

[25] Figure 3a presents regressions of dry filter measurements of Crahan *et al.* [2004] to dry APS estimated mass from 0.6 to  $10 \mu\text{m}$  diameter (which encompasses the principal salt mode while ignoring the artifact at the largest sizes). Included are gravimetric mass concentrations as well as a reconstructed sea-salt concentration from Na (utilizing the commonly used factor of 3.25, based on the makeup of seawater, for the conversion). The gravimetric regression include the 1:1 line with an  $r$  value of 0.82, but there is a slope offset of  $\sim 8\%$ . As expected the regression to sea salt is better ( $r = 0.88$ ) with a slope equal to the gravimetry line fit, but with a  $1.5 \mu\text{g m}^{-3}$  offset in concentration. From the PCASP data, most of this difference can be accounted for in the fine mode (below  $0.6 \mu\text{m}$  in diameter), ranging from 0.5 to  $1.5 \mu\text{g m}^{-3}$  with an average value of  $0.9 \mu\text{g m}^{-3}$ . Given the very large corrections required by Armendariz and Leith [2002] and other ambiguities such as density, dynamic shape factor, chemistry and inlets, we consider this comparison relatively good. Even after correcting for the fine mode, the gravimetry regression line is statistically not that different from the 1:1 line. However, there is subjectivity in our upper cutoff diameter on the APS and its relationship with the artifact at larger sizes (we need only to select an upper limit of  $8 \mu\text{m}$  to correct for the slopes). Aside from the obvious instrumentation issues, the slope difference in the sea salt versus the APS can in part be traced back to physical grounds as discussed later.

[26] Plots of 2-hour average dry number and volume size distribution for the R/P FLIP APS are presented in Figures 3b and 3c. Here we picked cases that corresponded to low wind and significant transported sea salt with a source region likely around tropical cyclones 3000 km away (4 and 10 September, respectively). Shifts in particle size to ambient diameters, are presented in Figure 3d. While particle concentrations changed greatly during the study, the overall shape of the size distribution did not significantly shift. A single mode dry volume median diameter of  $2.5\text{--}3 \mu\text{m}$  was generally present, with geometric standard deviations ranging from 1.6 to 2.0.

Hygroscopic growth of particles to ambient diameters (by approximately a factor of two) results in an ambient volume median diameter of  $\sim 4\text{--}5 \mu\text{m}$ , and a geometric standard deviation of 1.6. On the smaller end of the APS size spectrum, the application of the growth term results in the amplification of small perturbations (or “castling”). Given the narrowness of the size bins in the APS, such behavior is not unexpected.

[27] With its reasonable mass closure, and the APS’s nature as a single particle counter, it follows that the size distribution measurements for dry particles should also be reasonable. The third power relationship between number and volume is an extremely tight constraint on the system (even the smallest difference is quickly magnified in the volume distribution). These values also compare well to other APS measurements in the literature (Table 1). Given the general closure of the FLIP APS and filter measurements along with the physical consistency with other aerodynamic measurements in the field, in this paper we use this instrument as a secondary standard to compare all other size measurements. This is not to say that these measurements are necessarily “correct,” only that they are, in our opinion, the most reliable to proceed with in this intercomparison.

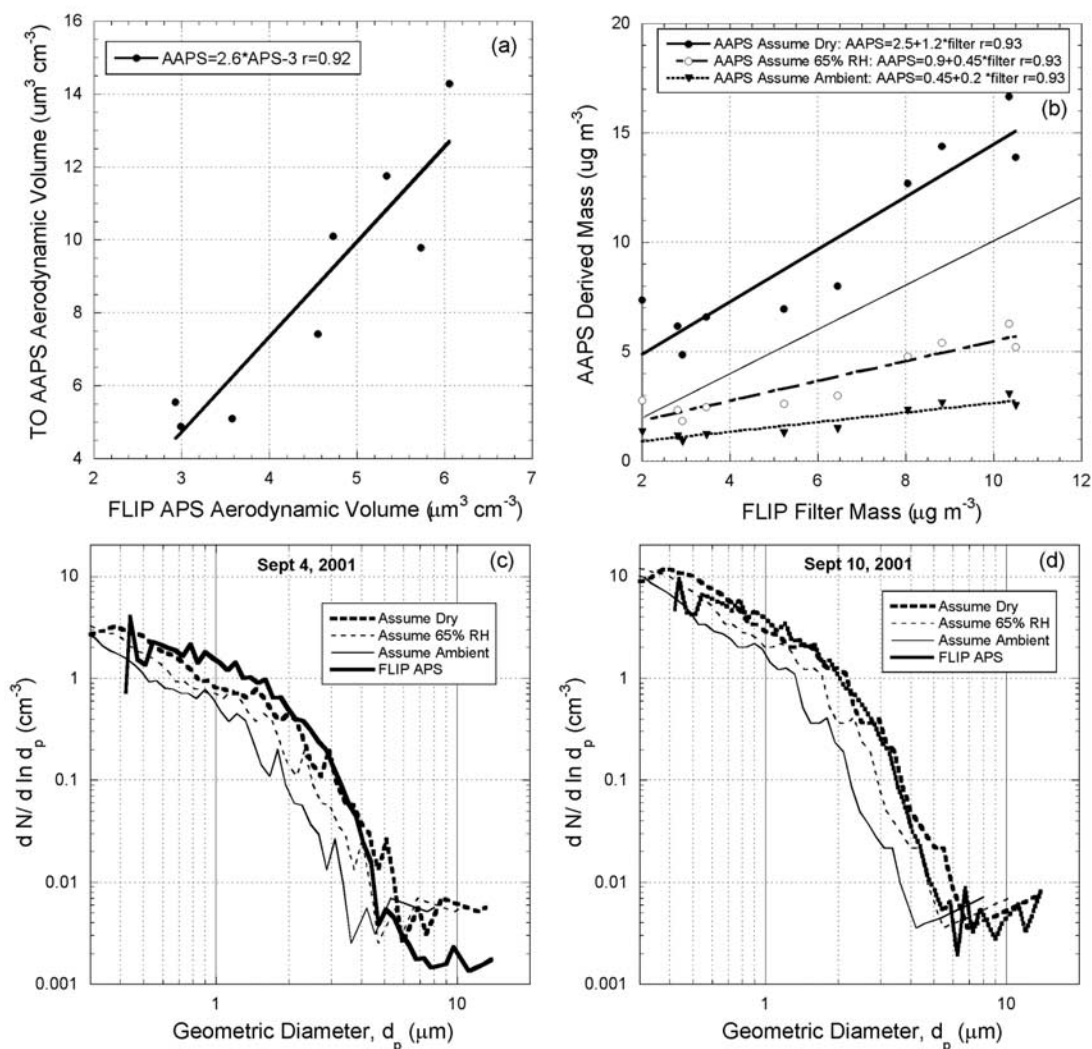
### 3.3. CIRPAS Twin Otter APS

[28] Because of the general success of APS instruments on shipboard vessels, CIRPAS sponsored the creation of an aircraft wing-mounted pod version using the core APS 3320 instrument (henceforth Aircraft APS, or AAPS). Despite both platforms using the core APS3320 instrument, the AAPS differs from the FLIP APS in a few respects. In the closure studies of Wang *et al.* [2002], it was estimated that the AAPS inlet cut point was on the order of  $8 \mu\text{m}$  (aerodynamic). However, this has been disputed and it maybe as low as  $\sim 4 \mu\text{m}$ . To date, neither of these numbers has been reproduced.

[29] The issue of relative humidity is critically important. The FLIP APS was essentially dry. However, the inlet to the AAPS is not heated, although some degree of drying likely occurs due to ram heating. This effect was estimated to be  $1.2^\circ\text{C}$  by Wang *et al.* [2002], or a 5% RH depression for the conditions we experienced in RED. However, this assumes no heat exchange between the particles, sheath flow, and the AAPS instrument itself. Further, flow through an orifice jet such as that on an APS is not adiabatic and results in a drop in relative humidity (H. Maring, personal communication, 2004). Hence RH of measured particles is neither ambient nor dry, but somewhere in between and is not measured in the instrument. This is not an uncommon situation, and is why researchers often heat the aerosol to ensure some known RH.

[30] To begin, let us assume that the AAPS yields a particle size distribution at an intermediate relative humidity of say 65% (compared to  $\sim 75\text{--}80\%$  ambient). This is approximately double the RH depression suggested by ram heating alone but within other observations (H. Maring, personal communication, 2004). The resulting AAPS volume distribution corrected to 80% RH for the 10 September over flight is included in Figure 2a. In this case, the comparison of distribution shape between the FLIP APS and the AAPS is remarkably good. Modal diameters, and even the  $10 \mu\text{m}$  anomalous inflection point are similar.





**Figure 4.** Comparisons of Twin Otter Airborne APS (AAPS) against *FLIP* parameters during flybys. (a) AAPS volume versus *FLIP* APS volume and (b) AAPS-derived mass versus *FLIP* filter mass for various assumed sampling humidity. Number distributions for (c) APS and (d) AAPS for the cases of 4 and 10 September 2001, respectively.

Differences include a bit more oscillation in the distribution curve, and a roughly 25% reduction in total volume concentration in the AAPS (which could be argued to be a result of a slightly lower assumed humidity, or by invoking vertical gradient arguments).

[31] The consistency between the surface and airborne probes can be examined in Figure 4 where we compare computed particle volumes, masses and number distributions during coincident flybys for particles ( $0.7 < d_p < 10 \mu\text{m}$ ). Consider Figure 4a with raw particle volume from the AAPS (using instrument aerodynamic diameter) and the *FLIP* APS (also using aerodynamic diameter). Here we find a generally strong correlation ( $r = 0.92$ ) and a slope of 2.6. Because the *FLIP* APS is “dry” and the AAPS is somewhere between dry and ambient, we expect the AAPS to yield a greater volume. However, if the AAPS operates with only a 5% RH depression as suggested by Wang *et al.* [2002], we would expect a difference on the order of a factor of 4 to 7.

[32] This point is reinforced in Figure 4b where the AAPS-derived coarse mode mass ( $d_p < 10 \mu\text{m}$ ) is plotted against filter-based sea-salt aerosol particle mass. Here we plot three values of AAPS-derived dry mass, assuming the measured size distribution is dry, at 65% RH, or at ambient conditions. The correlation coefficient between the AAPS and the filter mass is even stronger than that with the APS ( $r = 0.93$  versus 0.9). Because of the strength of the growth curve (roughly a factor of two in diameter between dry and 80% RH) we expect the factor of 8 difference between the assumed dry and assumed ambient regressions.

[33] We can explore the AAPS’s behavior further by comparing its derived size distribution to that of the *FLIP* APS. Figures 4c and 4d presents number distributions for the two instruments for the cases of 4 and 10 September 2001, which correspond to days of no production and of high concentrations due to advection (and a small amount of production), respectively. In these cases we do not expect a

vertical gradient of sea salt to form. Here, like Figure 4b, we adjust AAPS particle size to dry conditions based on the assumptions that the AAPS is in fact measuring a dry, 65% RH, or ambient size distribution.

[34] The number distributions from AAPS are inconsistent with the hypothesis that large vertical gradients in part cause differences between the two instruments. Even for the case of 4 September when no white capping was present, and hence there is a small vertical gradient in particle concentration, the differences over most sizes are large. The most interesting feature of Figure 4 is the crossover in size distribution in the 2–4  $\mu\text{m}$  range where the AAPS overtakes the APS. While for smaller particles the differences between the two instruments are a factor of two to three, the crossover and agreement in the distribution shape for the region of the volume mode is what ultimately leads to the favorable comparison of the instruments.

[35] From Figure 4, we are really left with only two possibilities. First, the inlet cut point for the AAPS is in fact closer to 4 or 5  $\mu\text{m}$ . If so, the favorable comparison in volume between the AAPS and the *FLIP* APS would be coincidental. The AAPS would have underestimated larger particles due to inlet losses, yet gained volume from the growth of smaller particles to larger sizes. We would then be left with a self-sustaining volume distribution. The second possibility is that the AAPS dries aerosol particles much more than had been previously assumed. In order to gain agreement, this must be very close to the efflorescence relative humidity of  $\sim 40\%$  (as is plotted in Figure 2b). This issue will be dealt with again when we compare particle vertical distributions.

### 3.4. Passive Cavity Aerosol Spectrometer Probe (PCASP)

[36] The PCASP is probably the most commonly used particle-sizing instrument on aircraft, and has been shown to be relatively reliable in closure calculations. The last few channels of the PCASP are often used to determine the presence or qualitative strength of the coarse mode. Like all closed celled instruments (such as the AAPS), internal relative humidity and inlet losses are always issues of concern. Indeed, some have argued that inlet losses for particles greater than 1  $\mu\text{m}$  can be significant for large perturbations in aircraft attitude [e.g., Haywood *et al.*, 2003]. Unpublished studies from CIRPAS suggest that ram heating and internal temperature increases due to sheath air also reduce the sample RH from ambient to almost dry level. To ensure RH values less than  $\sim 35\%$ , many investigators run the deicing heater on the PCASP to add a few more degrees of temperature and ensure a dry size distribution (for example, on the CIRPAS Twin Otter, UW C-131 and UW Convair 580, this was common practice). Strapp *et al.* [1992] found that with the heater on, the PCASP size distribution did not vary for a relative humidity above 90%. Typically the heater alone can increase air sample temperature by over 20°C (e.g., reduce sample from 75% to 24% RH at 25°C). In the case of the RED experiment, the Twin Otter PCASP deicer was on during the course of the study. On board the *FLIP*, the PCASP was ventilated without the use of a deicing heater (for fear of damaging the instrument during relatively low wind speeds). However, due to the internal electronics and pump temperatures, and the pres-

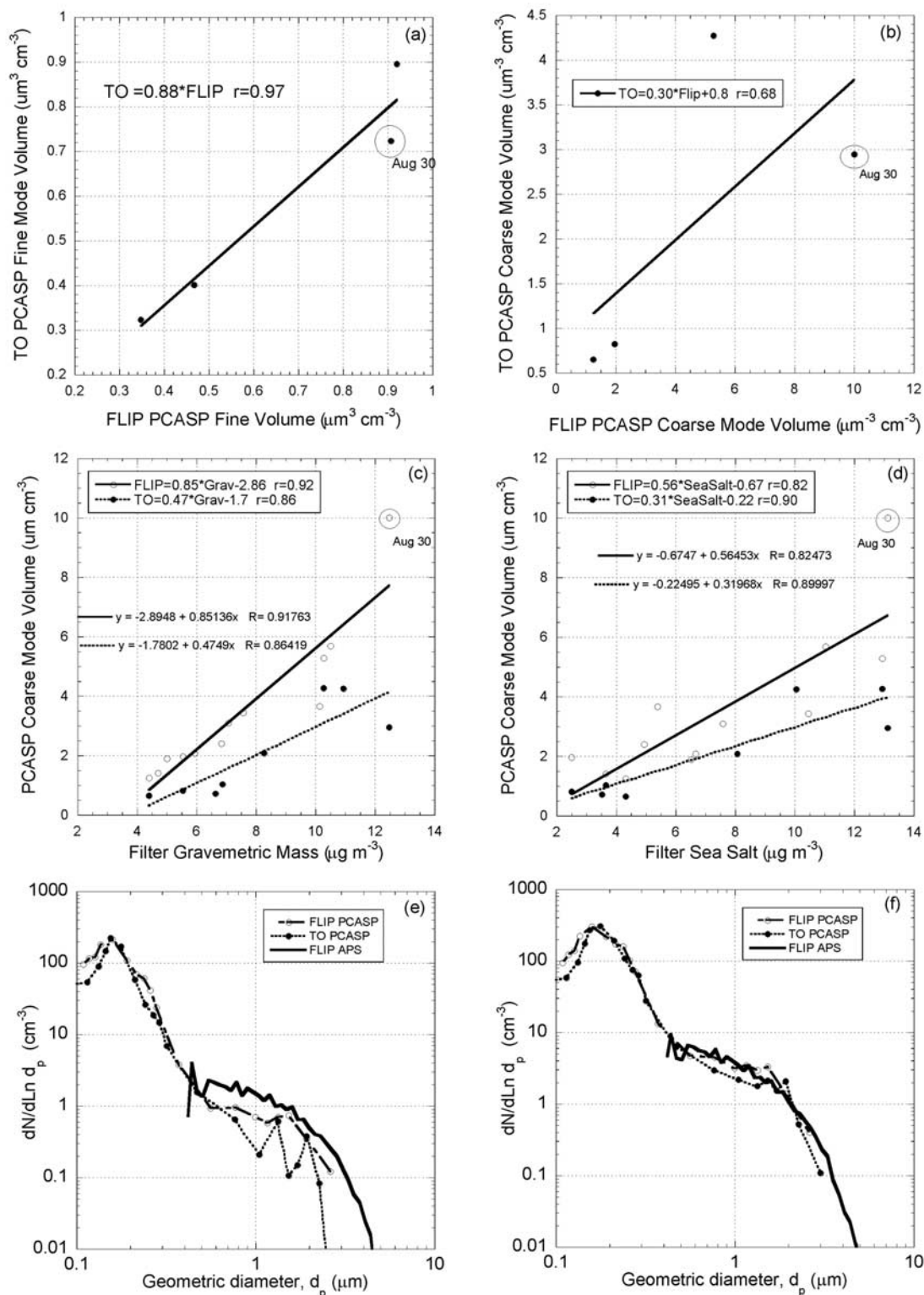
sure drop across the orifice we expect significant drying ( $\sim 10^\circ\text{C}$  or 40% RH).

[37] Over the course of the RED study, both the *FLIP* and Twin Otter PCASP instruments performed extremely well. In Figure 2a, where the PCASP volume distributions are shown for operating RH, the *FLIP* and Twin Otter instruments overlap for the bulk of the size range (up to 2  $\mu\text{m}$ ). An offset in volume relative to the APS on the order of 30–50% is related to our use of native RH, and can be reconciled by as little as 10% particle growth. Falloff in the Twin Otter PCASP after 2  $\mu\text{m}$  may be due to inlet issues. As the Twin Otter PCASP also had a deicer heater, we expected a smaller VMD in the fine mode as found (0.21 versus 0.22  $\mu\text{m}$ ), and a 10% smaller volume.

[38] Figure 5 presents intercomparisons of PCASP fine and coarse mode particle volumes, and their correlation with specific aerosol particle species. Here we use the curve minimum ( $\sim 0.6$   $\mu\text{m}$  diameter) to differentiate the two modes. An evaluation of the total response of the *FLIP* and Twin Otter PCASP in the fine mode is outside this study. However, for completeness it is worth discussing. On only four occasions did the Twin Otter and *FLIP* PCASP sample simultaneously, but the correlation was very strong ( $r = 0.97$ ) with three of the four points on the 1:1 line (Figure 6a). One point, corresponding to 30 August when there was active sea-salt production, was slightly offset from the others, and may be due in part to vertical gradients.

[39] The coarse mode of the PCASPs also exhibited reasonable behavior. Figure 5b shows the relative volumes of the Twin Otter versus *FLIP* PCASP. Here the correlation is not nearly as strong as for the fine mode ( $r = 0.68$  for 4 points). In this case, as expected, the outlier is again the 30 August case. To increase the number of samples, Figures 5c and 5d shows regressions of the two PCASPs versus gravimetry and sea salt, where correlations exist. The relationship between the *FLIP* PCASP and gravimetry is extremely strong ( $r = 0.92$ ), though 30 August remains an outlier. Since the *FLIP* PCASP and the filters ran at the same sampling height of 11 m, vertical gradient issues cannot explain these outliers. With respect to gravimetry and sea salt, the Twin Otter PCASP is smaller than the *FLIP* by about a further 30%. Such a difference may partly be due to additional drying. However, systematically throughout the data set, the last two PCASP channels of the Twin Otter drop off relative to the *FLIP* (as seen in Figure 2a). A large portion of this is likely due to inlet issues, calibration in the largest sizes or residual water.

[40] In the case of the *FLIP* PCASP, internal temperature probes were not installed, and though we expect a fair degree of drying to occur, the exact amount is unclear. However, our analysis indicated that the *FLIP* PCASP size distributions should be considered nearly dry,  $\sim 40\%$  RH near the efflorescent RH for salt ( $d_p/d_o = \sim 1.1$ ). Compared to other instruments (Figure 2b), the assumption that the PCASP is completely dry yields estimated mass values that are too high, on the order of 30%. Again, this is a very small hygroscopicity correction. In Figures 5e and 5f we present number distributions for the flybys of 4 and 10 September. In these plots we assume that both PCASP instruments are dry for index of refraction calculations. In this case, the comparison between the instruments is fairly good. For the *FLIP* PCASP, the number distributions completely overlap

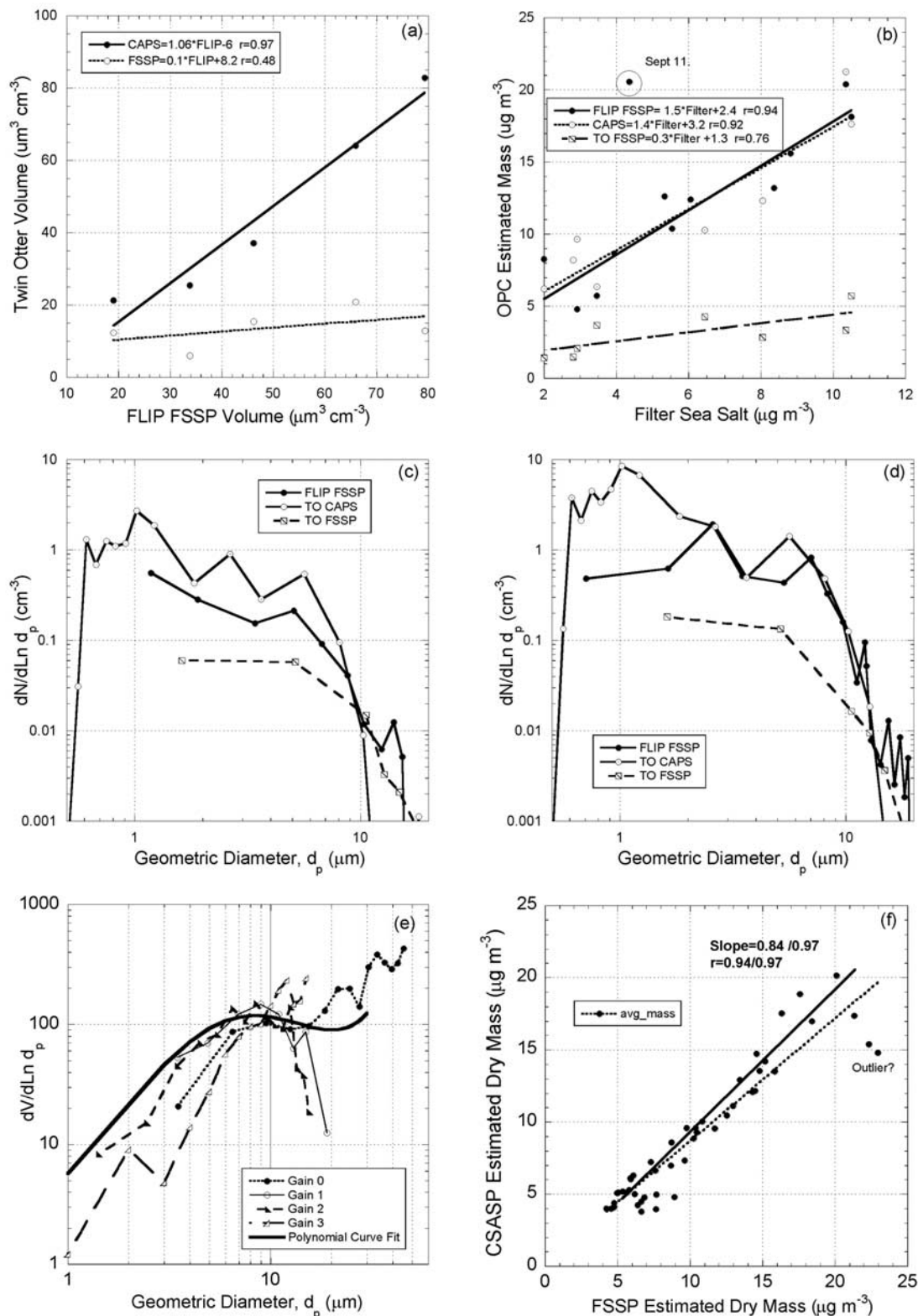


**Figure 5.** Intercomparisons of Twin Otter PCASP (a and b) coarse and fine mode volumes versus the *FLIP* PCASP, (c) filter chemistry, and (d) comparison of number distributions for the TO PCASP. (e and f) *FLIP* PCASP and *FLIP* APS for the cases of 4 and 10 September 2001, respectively.

the APS size spectra. On average, the Twin Otter PCASP runs  $\sim 20\%$  lower than the *FLIP* PCASP for diameters less than  $1 \mu\text{m}$ , where more divergence occurs. Interestingly, in the case of 4 September the divergence between the instru-

ments is somewhat greater, with oscillation evident in the Twin Otter PCASP. Examination of the data suggests that the particle concentration was so low during the flyby that there was not enough counting statistics for a proper





**Figure 6.** Comparison of Twin Otter and *FLIP* OPCs. (a) Twin Otter CAPS and low gain FSSP versus *FLIP* midgain FSSP. (b) Twin Otter CAPS, low-gain FSSP, and *FLIP* midgain FSSP versus filter sea salt. Comparison of number distributions for (c and d) the *FLIP* FSSP and TO CAPS and FSSP for the cases of 4 and 10 September 2001, respectively. Comparison of multigain stage CSASP with *FLIP* FSSP. (e) The 10 September case shown in Figure 2 with all gain stages and derived polynomial fit. (f) Comparison of CSASP gain 1 estimated dry sea salt mass concentration. Two sets of regression lines are shown. Solid line, all data; dotted line, exclude the two outliers from 30 August.

analysis. Regardless, these RH issues with the PCASP demonstrates the fair amount of subjectivity in deriving mass measurements.

### 3.5. Coarse Mode PMS Optical Particle Counters

[41] The PMS FSSP and its bench top derivatives, such as the CSASP, have been the mainstay of cloud and coarse particle research for nearly 30 years, and have undergone a number of wind tunnel calibration, intercomparison and sensitivity studies resulting in a number of corrections [e.g., Baumgardner *et al.*, 1990]. Despite the fact that the FSSP was designed and calibrated for cloud research, it has been consistently used in research aircraft for studying coarse mode particles. To this end, gain modifications and revised sampling protocols have been developed [e.g., Horvath *et al.*, 1990]. Bench top models of the FSSP converted for ground use for this purpose (e.g., CSASP-100HV and CSASP-200) have also been used. To derive particle size in the 1–10  $\mu\text{m}$  range in older versions of the FSSP and CSASP, a number of gain stages have been used (for example, 0.5–8  $\mu\text{m}$ , 1–18, 1.5–32, or 2.5–52 etc.). To extend the range of these types of OPCs, DMT developed the CAPS probe, which uses a logarithmic amplifier and measures particles from 0.5 to >50  $\mu\text{m}$  in one set of electronics.

[42] Figure 2a clearly shows the wide variability in coarse mode FSSP-derivative measured size distributions. As in the measurement of dust for PRIDE, these instruments result in VMDs (>6  $\mu\text{m}$ ) that cannot be reconciled with the filter or APS data (Figure 2b). Total volume is considerably higher, and at individual size bins, variances in particle concentration range over a factor of 3. Such differences were consistent throughout the entire study. The *FLIP* FSSP operating in a mid gain mode consistently yielded a VMD on the order of 9  $\mu\text{m}$ , with a broad shoulder extending to the end of its sizing range ( $\sim 22 \mu\text{m}$ ). Particles in the first two channels were underestimated by about a factor of 3. The CAPS forward scattering probe matched the *FLIP* APS extremely well to  $\sim 4 \mu\text{m}$ , after which it too formed an anomalous large peak (VMD =  $\sim 7 \mu\text{m}$ ). A similar shape (although smoothed) to the CAPS and FSSP was also found in the CSASP polynomial curve fit of its four gain stages. The Twin Otter FSSP, operating in low gain or “cloud” mode, underestimates channel one significantly and has poor fidelity out to 20  $\mu\text{m}$ .

[43] Despite the sizing differences between the coarse mode FSSP-derivatives and the APS, the CAPS and *FLIP* FSSP track one another reasonably well (Figure 2b). Figures 6a and 6b present regressions intercomparing these instruments with other available data. In Figure 6a, we find a near perfect match between the Twin Otter CAPS and *FLIP* FSSP volumes during the 5 overpasses when the instruments were simultaneously operating (slope = 1.06,  $r = 0.97$ ). The Twin Otter FSSP however, did not show so good a response (slope = 0.1 and  $r = 0.49$ ), likely due to its limited channels in the region of the sea-salt mode.

[44] Estimated dry particle mass is compared to filter mass in Figure 6b. Here the Twin Otter FSSP and CAPS also have a strong correlation with the filter data ( $r = \sim 0.93$ ), although over sizing results in a  $\sim 50\%$  overestimate of dry particle mass. This is consistent with APS data as well (not shown). Here we have removed the outlying

datum point for 11 September, where the filter dramatically underestimates Na (from which sea salt is derived). However, if we used particle gravimetry, this data point would move down to join the others (this point is also an outlier relative to the PCASP as well). Regressions for the Twin Otter FSSP to dry sea salt are better than its comparisons to the other two probes, but total dry volume is still underestimated by  $\sim 50\text{--}70\%$ .

[45] In Figures 6c and 6d we compare the number distributions of the *FLIP* FSSP with the CAPS and Twin Otter FSSP. Like previous comparisons, we use 4 and 10 September because of a low probability of vertical gradients. Here too we find consistent differences. Each of the instruments clearly undercounts in their first two channels. Particle undercounting in these instruments in their first channel is well known to the community [e.g., Horvath *et al.*, 1990]. As boundaries are ill defined, it is a matter of course to throw out the first channel in any OPC (this procedure is performed at all of the major research aircraft operations). However, it appears that one should also limit the use of channel 2 and modify 3 because particles are still near the signal to noise threshold, and the sampling volume is ambiguous.

[46] In the largest sizes, oscillation in the spectrum is also evident. This can be simply attributed to poor counting statistics. Even so, for particles greater than 12  $\mu\text{m}$ , the CAPS probe is consistently lower than the other two FSSPs. Because the Twin Otter is over the *FLIP* for less than a minute, this too could be a sampling issue.

[47] What is most dramatic is the difference between the instruments in the 1 to 10  $\mu\text{m}$  range, where the bulk of the sea-salt mass exists. Here we see quite a bit of oscillation in the instruments’ size curve. However, because the volume distribution is tightly forced around 7–10  $\mu\text{m}$  (Figure 2a), this behavior does not seem to affect the comparisons of these probes with dry particle mass. Also, it is clear that the outstanding comparison of particle volume between the CAPS and the *FLIP* FSSP is, to a large degree from cancellation of errors, with the smaller sizes of the CAPS making up for the increased volume in the FSSP for  $d_p > 10 \mu\text{m}$ .

[48] Also onboard the *FLIP* was the TNO CSASP, the bench-top derivative of the FSSP. The CSASP is similar in geometry to the FSSP, but with its own ventilation system and a short sampling horn. While the CSASP should behave very similarly to the FSSP, it does have the ability to operate in 4 gain ranges simultaneously. The four stages pose a problem; how are they best utilized in analyses? Some investigators have subjectively included or removed stages based on the assumed size of the aerosol. In the case of the data collected by TNO, all stages are used to generate a fifth-order polynomial fit.

[49] The TNO CSASP operated for nearly the entire study and in general behaved similarly to the boom mounted FSSP. Figures 6e and 6f presents representative data for the mission (a complete analysis by de Leeuw *et al.* is forthcoming). To reiterate, these data were generated using the same calibration and analysis procedures used by TNO in all of their previous studies. Hence it is a data set independent from the NRL and CIRPAS investigators. To begin, consider Figure 6e where the individual gain spectrums from the 10 September event are presented along

with the polynomial fit. While the individual spectra can vary considerably for each size region, most show fairly similar characteristics. The anomalous  $8\text{ }\mu\text{m}$  peak is clearly evident in each stage, as well as the polynomial fit. The first few channels in each stage are considerably lower than the others. Significant drop-offs are also visible in the last few channels of some of the stages. Despite this, the middle channels for each stage are fairly consistent. Such behavior is very similar to the spectra given in other multistage OPC such as the ASASP (e.g., see the collected works of Pinnick).

[50] While the basic shape of the CSASP is very similar to the *FLIP* FSSP, interpretation is not straightforward. Consider Figure 6f, where we present the regression of estimated dry mass between the CSASP gain stage 1 (most closely resembling the channel sizes of the *FLIP* FSSP) and *FLIP* FSSP (6 hour average). On the whole, the two instruments track extremely well ( $r$  values above 0.95) with near unity slopes. Two regression lines are also given for regressions that include and remove the two largest outliers corresponding to 30 August (where vertical profile may be important, since the CSASP was 8 m higher than the FSSP). Because gain stage 1 and the FSSP agree so well, we are reasonably confident that the two instruments were operating and calibrated as they were intended. However, interpretation for multiple gain stages becomes much more problematic. Estimated mass concentrations can vary by more than a factor of 3 based on individual gains, or how they are weighted into a continuous curve fit.

[51] The CAPS, FSSPs and CSASP described here exhibit overestimation of sizes with an unphysically large VMD in the 6 to  $9\text{ }\mu\text{m}$  range, and a small geometric standard deviation of 1.5. This is a result of a fairly consistent peak (or at best a flattening) of the size spectrum. The issue of particle over-sizing in the coarse mode OPCs seems to be chronic. The optics of the FSSP derivatives and the forward CAPS probes are very similar. Hence it is not surprising that they all exhibit over sizing (and hence yield overestimates of volume). This over sizing and resulting overestimate of particle volume is similar to the findings of Collins *et al.* [2000] and Reid *et al.* [2003] for airborne dust, and is certainly due to the degeneracy in the particle size-voltage response function for these instruments. Plots of this response function are presented in both of the above manuscripts.

[52] Lengthy discussions as to the nature of the response curves of these instruments can also be found in the references above; these curves are expected to be particularly a significant problem for airborne dust. Fundamentally, optical particle counters use a measurement of light scattered off of a particle from a white light or laser beam into a detector. Voltages from the detector are related to those from some standard sphere of known size and index of refraction. Simply put, these instruments measure pulse heights in a detector, and relate that pulse height to some empirical curve fit using standard spheres. Almost all investigators correct these calibration curves to account for the mean index of refraction of the aerosol particle type being measured, as we have done. However, as is evident, the FSSP and CAPS probes are insensitive to particle size in the  $\sim 3$  to  $10\text{ }\mu\text{m}$  range. This is due to frequent Mie oscillations, and compounded by an inflection point at  $\sim 5\text{ }\mu\text{m}$  in the Mie

size-scattering cross section curves at the FSSP and CAPS scattering angles. As particle index of refraction increases, instrument sensitivity decreases and the degeneracy worsens. Ultimately, some mean calibration curve is drawn. The strength of the degeneracy is weaker for particles with lower refractive indices (such as hydrated sea salt), and typically the scientific community simply draws a one-to-one curve that was thought to overcome this problem. Clearly from this and other work, such a procedure does not address the fundamental physics problem. Since volume goes as the radius cube, it does not require missizing of many particles to significantly bias the volume distribution.

[53] In addition to the degeneracy bias, there is clearly also a gain bias in the FSSP derivatives that is less publicized. Each of the gain stages of the CSASP tends to exhibit the same biases as the FSSP with significant drop-outs in the first channel. The inconsistent nature in the lower gain stages of the CSASP (i.e., larger particles) is more troubling and we do not have a clear causal reason other than “issues” with the electronics (the CAPS system however, with its large single gain stage seems to be immune from this problem). This behavior in the CSASP is similar however to that seen in intercomparisons of the FSSP-100 (i.e., low gain) and FSSP-300 (high gain) instruments on the UW C-131 during the SCAR-B campaign [Reid, 1998]. In that study, the FSSP-100 presented an extremely large coarse mode that was not seen at all in the FSSP-300. However, both instruments compared well in their mid range. This bias is particularly troubling since it makes the estimation of particle size errors for the largest particles highly dependent on gain.

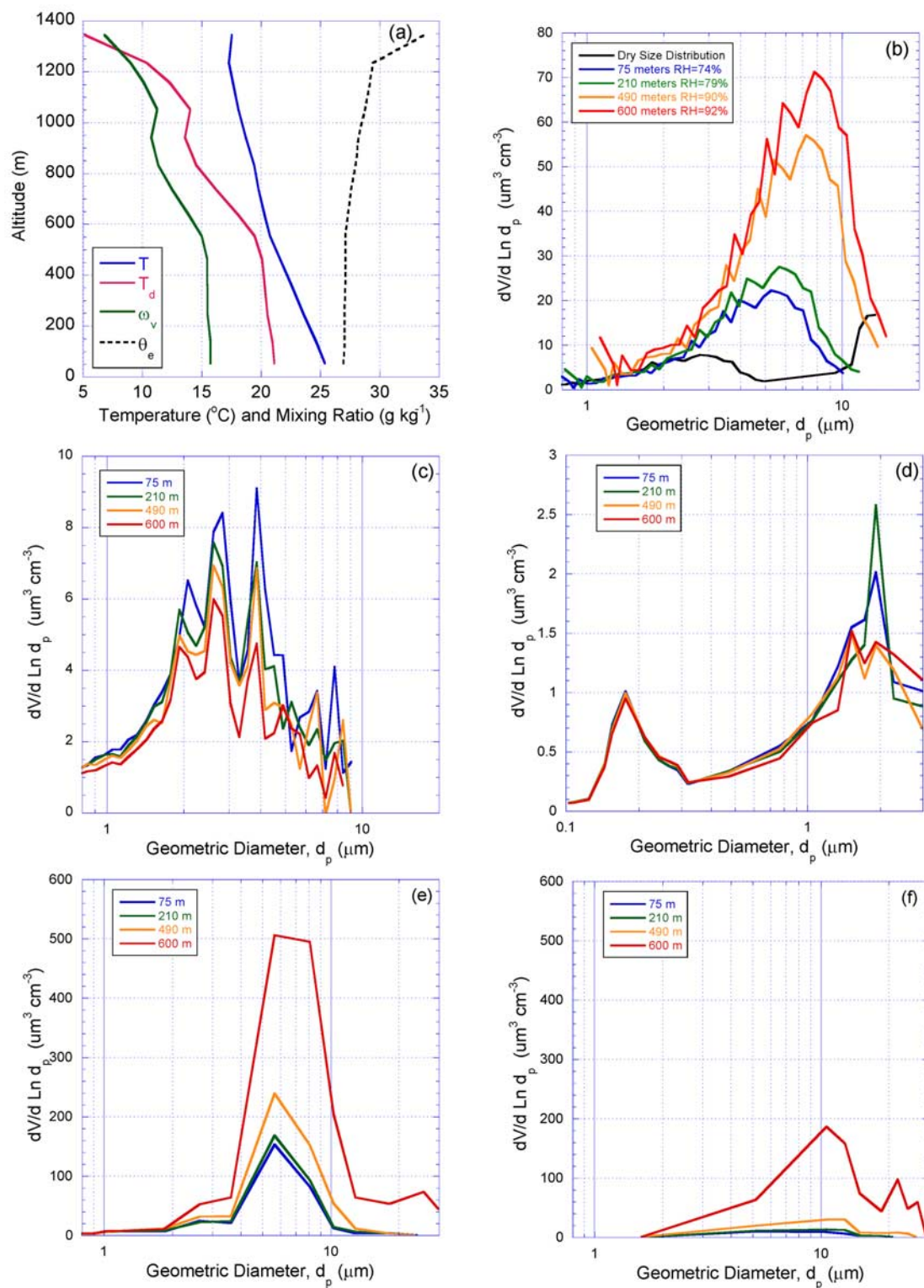
### 3.6. Examination of Particle Vertical Distribution

[54] The advantage of the AAPS, FSSP and CAPS probes is that as they are wing mounted, so one should be able to derive information on the vertical profile of the aerosol particles. While a full evaluation of sea-salt vertical profiles is outside the scope of this manuscript, there are some fundamental issues worth examining. We have shown that all probes at least track particle concentrations reasonably well. Also, during the *FLIP* flybys the shape of the particle size distribution did not appear to appreciably change at the surface. Given that the CAPS, FSSP, and AAPS have known biases, to what extent can they capture shifts in the particle size distribution? In particular, we should be able to observe a shift to larger particle size with altitude in the MBL due to hygroscopic growth. To what extent do these high RH values affect size spectra interpretation?

[55] We begin our analysis with 10 September, a day with little cloud cover and a fairly straightforward boundary layer structure where the relative humidity ramps from  $\sim 74\%$  near the surface to  $90\text{--}92\%$  in the vicinity of the inversion (Figure 7a). In the upwind region of the study area (approximately  $100\text{ km}$  offshore), the Twin Otter performed a series of 10 minute, level altitude, cross wind legs throughout the marine boundary layer. Data in Figure 7 are density corrected.

[56] Figure 7b presents an ideal case where we have taken the dry size distribution from the *FLIP* and grown the size distribution to the respective RH of the levels assuming a





**Figure 7.** The vertical distribution of particle size in the marine boundary layer above *FLIP* for 10 September. (a) Thermodynamic sounding of profile. (b) *FLIP* APS with inferred size distributions using the Gerber [1985] growth factors. (c) Twin Otter AAPS, (d) Twin Otter PCASP, (e) Twin Otter CAPS forward probe, and (f) Twin Otter low-gain FSSP.

well-mixed MBL. Here we find the distributions grow smoothly from a VMD of  $\sim 3 \mu\text{m}$  for dry conditions to  $\sim 8 \mu\text{m}$  at 92% RH.

[57] In the case of the wing mounted AAPS (Figure 7c), we find that the steep oscillation in the curve is consistent throughout the vertical profile, likely due to minor bin width errors and should be of little concern. More important however, is the finding that the volume concentration goes down with increasing altitude and RH. If the AAPS gave a strictly dry size distribution, the mixing in the MBL would result in static size spectra. If the AAPS were close to ambient, it would increase in amplitude similar to Figure 7b. Here we find a 25% reduction in particle concentration at  $1 \mu\text{m}$  in diameter between 74 and 92% RH. This increases to a 40% reduction for  $3 \mu\text{m}$  particles and more than a factor of two for  $4 \mu\text{m}$  particles. The size spectra then become jumbled for particles greater than  $5 \mu\text{m}$ . This reduction in concentration with increasing RH leads to suspicion of the inlet design. The largest falloff, in the  $3\text{--}6 \mu\text{m}$  range, is consistent with sampling losses. The deviance in the  $\sim 1 \mu\text{m}$  range is not as great, but is statistically significant. It could be argued that at the higher RHs, particles in this range have grown to a larger size where inlet losses are greater. We find this hypothesis possible, but unlikely. The geometric growth of particles from 74% to 92% RH is only about 45%. Another possible cause is that the AAPS suffers from other internal losses inside the instrument itself that are exacerbated at the higher RH values.

[58] Figure 7d displays particle volume spectra from the PCASP. The clearly static nature of the accumulation mode during the profile in the well-mixed MBL demonstrates the high degree of drying in a PCASP with its heaters on. Only a minor deviation ( $<10\%$ ) was found in the  $0.26\text{--}0.3 \mu\text{m}$  range. Significant differences however are visible for particles greater than  $\sim 1.5 \mu\text{m}$ . Like the AAPS, we find a reduction in particle concentration with increasing RH for particles greater than this size. In this case however, the combined particle growth/inlet loss hypothesis is probably valid. Here there is more than a factor of 2.5 difference in size between the dry and ambient particle size. Given that the inlet for the PCASP was designed with a  $4 \mu\text{m}$  cut point to begin with, one should probably not expect any better performance. Even so, it is a good reminder of the limits of coarse mode interpretation with the last few channels of the PCASP.

[59] The CAPS probe (Figure 7e) and the low gain FSSP (Figure 7f) respond to changes in RH during the vertical profile. However, this growth deviates significantly from theory. While these instruments yield spectra of increasing volume with RH (as they should), it is clear that the VMDs are not increasing with them. For both the forward CAPS and FSSP, the size spectra seem almost static in shape up to 90% RH, with some change only being seen at 92% (similar to the findings of Reid *et al.* [2003], who found static distribution shapes in the measurements of coarse mode particles through an inversion). The rate of increase in particle volume also deviates significantly from what we expect. The increase in volume from 74 to 92% RH is similar between the APS theory and forward CAPS (a factor of  $\sim 3.5$ ). However, the 90% RH leg at 490 m is smaller than we would expect. Similar behavior can be seen for the FSSP, although the VMD is slightly larger. Ironically, at the

highest relative humidity, the VMD from the PMS probes are serendipitously close to reality.

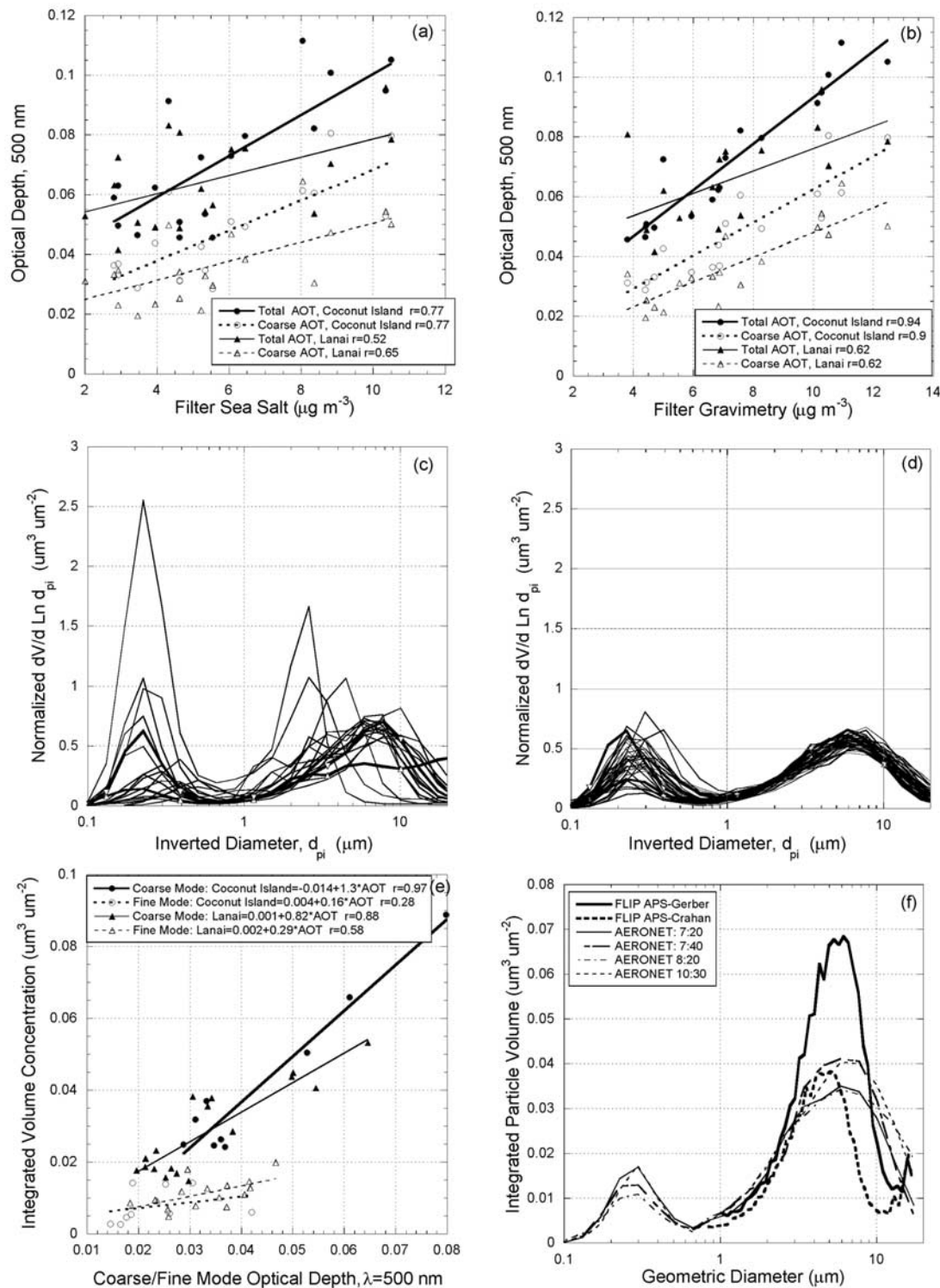
[60] The likely reasons for the behavior of the forward CAPS and FSSP relate to the same response function issues discussed in the previous section. In the case of hygroscopic growth in the MBL, high concentrations of smaller particles grow into the degeneracy range. These consequently have a higher contribution to the over-sizing problem. The relative biases in the volume distribution are then dependent on the shape of the original aerosol particle size spectra. In the case of the forward CAPS, the end of the major inflection point is approximately  $6 \mu\text{m}$ . In this case, particles are growing into the bin in which they were originally oversized. Consequently, there is not as large an increase in volume for RH values up to 90%. However, in the case of 92% RH, particles are now physically growing into the next bin, and the increase in volume and VMD is evident. A similar argument can be made for the FSSP. In its case however, we still must account for the chronic undercounting of the first two channels, which results then in a VMD of  $11 \mu\text{m}$  instead of the  $6 \mu\text{m}$  for the forward CAPS.

### 3.7. Comparison to Sun/Sky Inversions

[61] Retrievals of particle volume distributions and optical properties using the Sun/sky inversion method of Dubovik and King [2000] are increasingly finding their way into models and analysis. The Dubovik inversion uses a combination of spectral AOT and almucantar sky radiances at 440, 675, 870 and 1020 nm to invert particle size distribution and index of refraction, from which other optical parameters such as single-scattering albedo and asymmetry parameter are derived. Contrary to popular belief, at no time does the inversion force the presence of modes, although it does require a slight amount of smoothing (e.g., no casting between individual bins). A complete discussion of the application of this and other retrievals to coarse mode dust is discussed fully in the work of Reid *et al.* [2003]. Much of the analysis is applicable here, though in the interest of brevity, interested readers are referred to the manuscript.

[62] Typically, inversions are applied to periods of moderate optical depth. In the case of the marine environment, optical depths are extremely low and even small calibration errors in the Sun photometer can have significant effects. This is particularly true with respect to index of refraction and absorption properties. However, size distribution retrievals are more robust because they are derived from the relative angular distribution of sky radiance rather than absolute values. Hence sea-salt climatologies using inversion methods have been developed and applied in the scientific community to size distributions and the asymmetry parameter [e.g., Dubovik *et al.*, 2002; Smirnov *et al.*, 2003]. Because these inversions give output on column-integrated values at totally ambient conditions and are constrained to total optical depth, they are often preferred by the remote sensing and radiation communities. In our analysis of sea-salt particle size distributions, we examined the retrieved size spectra from the nearby AERONET site on Coconut Island, Kaneohe Bay (15 km away) and the island of Lanai ( $\sim 100$  km away).

[63] Optical depths at 500 nm at the Coconut Island site varied from 0.05 to 0.12 and, based on the coarse fine



**Figure 8.** Comparison of AERONET optical depths and inversion parameters. (a) Coconut Island and Lanai total and coarse mode AOTs versus *FLIP* sea-salt concentrations from chemistry. (b) Coconut Island and Lanai total and coarse mode AOTs versus *FLIP* sea salt concentrations from gravimetry. (c) Normalized level 1.5 volume distributions from Coconut Island for the RED study period. (d) Normalized level 2.0 volume distributions from Lanai for the RED study period. (e) Integrated volume concentration versus the coarse and fine mode optical depths. (f) Column-integrated particle volume distribution from AERONET inversions and those inferred from the surface-based APS.



partition from the technique of *O'Neill et al.* [2001], we estimated the coarse mode particles account for  $\sim 2/3$  of the total (e.g., Figure 1). This increases to  $\sim 90\%$  at 1020 nm. The coarse mode optical depth generally tracked with particle concentrations on the *FLIP*. This qualitative agreement was also observed with the island of Lanai, although coarse mode particles only account for 50% of optical depth at that site.

[64] Figure 8 presents various regressions and size distributions from the Coconut Island and Lanai AERONET sites. In Figure 8a both total and coarse mode optical depth are plotted against dry sea-salt concentration at the *FLIP*. Despite the fact that optical depth is an integrated quantity, the regressions are relatively good, with correlation coefficients on the order of 0.77 at Coconut Island and  $\sim 0.6$  for Lanai. While one might assume that much of the scatter relates to such parameters as humidity and boundary layer height, we found that some of this uncertainty is in the Na measurement from which sea-salt mass is derived. Indeed, like many other comparisons with instruments in this manuscript, the correlation is stronger against gravimetry (Figure 8b) that can account for contamination from the islands or Kilauea volcanic emissions. Part of the good correlation is no doubt to the relatively stable marine boundary layer depths in the subtropics.

[65] As one would expect, correlations are weaker at Lanai, some hundred kilometers away. Interestingly, however, there is a clear difference in the regressions. If one considers only the coarse mode optical depth, the bias is on the order of 30%. Given that the published uncertainty in AERONET AOTs is on the order of 0.01–0.015 [*Eck et al.*, 1999], some of these differences may be simply due to calibration. The difference in AOT trends at the two sites is almost certainly due in part to orographic forcing in the region. Lanai is often in the lee of Maui, and consequently sea-salt concentrations may be lower (J. Porter, personal communication, 2003). Further, Lanai is more likely to be influenced by output from the Kilauea volcano. The Coconut Island site has no upwind obstructions, but is extremely close to the windward mountains of Oahu. This mountain range causes in orographic lifting several kilometers out to sea, increasing the MBL height [*Porter et al.*, 2003]. This in turn increases the mean and maximum relative humidity of the MBL, and produces windward clouds. These clouds, while screened in the AERONET protocols, do reduce the number of points usable in an almucantar scan.

[66] The orographic effects at the Coconut Island site strongly reduce the applicability of inversion data there. In fact, the site is considered by AERONET staff to be one of the worst for retrievals in the AERONET network. During the RED campaign, the Dubovik retrieval was run on 18 cases at level 1.5 (e.g., preliminary retrievals). However, only two of these actually passed quality assurance to go to level 2 (final cloud screened and quality assured data). In the other 16 cases, the sky radiance measurements were fairly inhomogeneous. Level 1.5 retrievals normalized to coarse volume (for comparison) are plotted in Figure 8c, with the two level 2 retrievals in bold print and point markers. As can be seen, there is tremendous variability in both the fine and coarse mode, with volume median diameters ranging from 2.4 to 10  $\mu\text{m}$ . Distributions in the coarse mode are at times relatively jagged, similar to

those measured during the PRIDE campaign when cloud interference was suspected [*Reid et al.*, 2003]. Even the two retrievals that passed to level two have suspicious characteristics.

[67] Clearly, retrievals from Coconut Island cannot be (and have not been to our knowledge) used in any scientific study. However, the Lanai site is commonly used by the AERONET group and others to study the marine environment. During the RED field campaign, the Lanai AERONET site performed 35 successful level 2 retrievals, including 28 while the *FLIP* was deployed. In all of these cases, the solar zenith angle was greater than 30 degrees and Sun/sky errors were typically  $\sim 6$  and 3%, respectively. These conditions are within the bounds of required conditions for a “good retrieval” by *Dubovik and King* [2000]. Figure 8d presents these 35 volume distributions (also normalized). Unlike for the Coconut Island site, retrieved size distributions of the coarse mode at Lanai were much smoother. Distribution VMD averaged  $5.6 \pm 0.2 \mu\text{m}$ , with geometric standard deviations on the order of  $2.0 \pm 0.2$ . Variability in the fine mode size spectra was also smaller, with a fair amount of modulation due to pollution from Maui and/or output from Kilauea volcano.

[68] Figure 8e presents correlations of daily average integrated fine and coarse mode volume from the Dubovik inversion, versus fine and coarse mode AOT from the independent *O'Neill* algorithm, respectively. Coarse mode volumes correlated with coarse mode AOT relatively well at both the Coconut Island and Lanai sites. In the fine mode, correlations were weaker. Plots such as this tell much about the performance of an inversion. First, this is in part expected since there is more dynamic (and a much larger range) in the coarse mode sea salt due to wind speed and air mass history than for the fine mode. Further, the mass extinction efficiency for coarse mode particles, such as sea salt, is linear in VMD (e.g., see discussion in the work of *Reid et al.* [2003]). Hence any shift in particle size for a given fixed volume should be visible in a scatterplot such as this. In the case of Lanai, a reasonable regression exists and, given that the size parameters were fairly static, we expected such a result ( $r = 0.88$ ). However, the fine mode has a poorer correlation ( $r = 0.58$ ). In cases of low optical depth such as this, the retrieval likely focuses the constraint on the dominant term, in this case sea salt. Scattering by sea salt is predominately in the far forward, and hence the instrument shows good signal to noise. Conversely, fine mode particles predominately make their presence known through larger scattering angles ( $\sim 45$  degrees and larger) and the geometry of the measurement, along with their more diffuse scattering properties, results in poorer signal to noise. Because of mass scattering efficiency's strong size dependence in the fine mode, there is likely some degeneracy in the volume/size relationship. In addition, when one considers that the retrieval is attempting to estimate size on a fine mode optical depth of 0.02 to 0.04, the impact of this becomes clear. In order to provide the necessary optical depth constraint, the weaker fine mode likely suffers. Indeed, the sensitivity studies of *Dubovik and King* [2000] show that uncertainties in the total volume in these situations are likely on the order of 30%.

[69] Given that Coconut Island retrievals did not even pass internal AERONET quality control, and since the

aircraft could not fly directly above the site due to air traffic issues, we cannot perform a “column closure” comparison with measurements onboard the Twin Otter. However, we can compare the retrieved size distributions at Lanai with field measurements, and determine if the results are at least reasonable. For this comparison, we used the *FLIP* APS and two hygroscopic growth curves [Gerber, 1985; Crahan *et al.*, 2004]. Figure 8f presents a modeled, size column averaged, size distribution based on *FLIP* APS data, the relative humidity profile, and qualitative particle concentration from the Twin Otter for 1030 HST 10 September 2001. Also shown are four retrieved size distributions for the same day taken at Lanai. In this case, we corrected retrieved size distributions to match the 1020 nm AOT at Coconut Island (i.e., to help normalize the coarse mode). These corrections were on the order of 25%.

[70] Overall the shape of the retrievals, and the APS modeled column-integrated size distributions are close (APS VMD = 5  $\mu\text{m}$ , retrievals = 5.55  $\mu\text{m}$ ). The APS is somewhat narrower, with a geometric standard deviation of 1.65 versus 2.0 for the retrievals. However, there is a distinct amplitude difference between the model and inversions (on the order of a third) if one assumes the Gerber [1985] form of the aerosol hygroscopic growth curve. On the other hand, if we use the Crahan *et al.* [2004] form, we get a slight underestimate of particle VMD. This demonstrates how important it is to use correct hygroscopic growth functions. Regardless of the growth curve used, the retrieval has a significantly higher value of particle volume for diameter greater than 6  $\mu\text{m}$ . This may be due to those particles’ small optical cross section or the presence of larger particles aloft.

#### 4. Analysis and Reconciliation of Measurements

[71] In this manuscript we have intercompared particle size distributions and concentrations for some of the more commonly used methods in the field. Qualitatively, all methods track reasonably well, although significant differences in amplitude and size exist. In the following subsections we analyze each group of instruments separately, and attempt to reconcile our findings with those in the literature. It is said, “A man with one watch knows what time it is; a man with two watches is never sure” [anonymous]. In our case, there have been over a hundred relevant papers on the nature of sea-salt particle size and concentration. As will be shown, our findings are consistent with the bulk of these manuscripts.

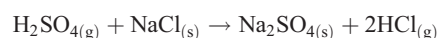
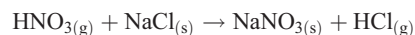
##### 4.1. Filter Gravimetry and Chemical Measurements

[72] From the start of this analysis, we choose to begin with one “benchmark” from which all comparisons would follow. We postulated that filter measurements taken aboard the *FLIP* would be the best standard, although this must be taken on a certain degree of faith. However, like the instrumentation discussed here, filter methods have their own set of issues including inlets, cut points, chemical speciation uncertainties, background subtractions, and gravimetric biases. Filter samples and gravimetry are nevertheless more fundamental than any of the sizing methods. In our case, we want to ensure that there are not any significant biases relative to the community data set.

[73] We do have a fairly high degree of confidence in the Na and gravimetric filter data. Our values are well within those reported in the literature before. Henintzenberg *et al.* [2000] found that based on chemical measurements in the literature, the mean sea-salt concentration for the 0–15° N, 15–30° N, and 30–45° N zonal regions were 8, 13, and 6  $\mu\text{g m}^{-3}$ , respectively (for comparison, our measurement latitude was 21°N). While low in comparison to this annual zonal mean, our 10 m s<sup>−1</sup> wind speed values are close to many other studies including Bressan and Lepple [1984], Marks [1990], Taylor and Wu [1992], and the results summarized by Quinn *et al.* [2000].

[74] On the basis of the PCASP fine mode background concentrations on the order of 0.5–1.5  $\mu\text{g m}^{-3}$  (from assumed density of ammonium sulfate/bisulfate of 1.8 g cm<sup>−3</sup>), coupled with the Na\*3.25 method to determine the mass of sea salt, we do have reasonable mass closure. Because the Na\*3.25 method is based on the stoichiometry of dissolved ions in seawater, it likely underestimates the total amount of “dry” mass for several reasons. First, Tang *et al.* [1997] found that for “dry” sea salt (as opposed to simple sodium chloride); there is still a possible 10–20% weight bias below the efflorescence RH (~35–40%) due to tightly bound water. This water is almost impossible to outgas under typical sampling and analysis conditions [e.g., Tang *et al.*, 1997]. Consequently, it is included in the gravimetric determination of aerosol particle mass concentration, but decreases the “effective” density of the particles by ~5%.

[75] A second issue relates to chlorine depletion. During the sea-salt aging process, gas phase H<sub>2</sub>SO<sub>4</sub> and HNO<sub>3</sub> react with NaCl, producing particulate sodium sulfate/nitrate and out-gassing HCl:



Because Cl has an atomic weight of 34, and nitrate and sulfate have molecular weights of 62 and 96, respectively, each release of Cl results in a further increase in the ratio of sea-salt mass to Na. For each 10% of Cl mass depletion, the ratio of sea-salt mass to Na increases by 2% or 4% for the nitrate and sulfate reactions, respectively. For the RED experiment, the filter data yielded a 5 to 40% Cl mass depletion (lower depletion for the high wind event on 30 August and 1 September). This suggests a negative bias in the sea salt to sodium ratio of up to another ~15%. As found by de Leeuw *et al.* [2003b], Cl depletion by nitrate can be rapid, especially near coastlines and this effect must be taken into account. Again, like tightly bound water, this bias is accounted for in our gravimetric analysis.

[76] Lastly, the use of a sodium indicator for sea salt does not account for any nonvolatile or semivolatile organic species. While values as high as 20% to 50% have been suggested [e.g., Novakov *et al.*, 1997; O’Dowd *et al.*, 2004], values on the order of 10–20% are certainly accepted [Crahan *et al.*, 2004]. In the case of semivolatile organics, it is unlikely that they are fully accounted for in the gravimetry. However, since these semivolatiles are in equilibrium with the surrounding atmosphere, they are probably measured by particle probes.

[77] Given all of the above considerations, one cannot expect better than  $\sim 15\%$  mass closure based on the  $\text{Na}^*\text{3.25}$  method and gravimetry, and negative biases as large as 10–25% are still possible from filter methods in totality. Hence these should be considered the baseline uncertainty from a well sampled air stream, and the APS filter relationships are about as good as one can expect under field conditions. While these uncertainties can seem high, as shown these are relatively small in comparison to other methods.

#### 4.2. Hygroscopic Properties of Coarse Mode Sea-Salt Particles

[78] The bulk of the field data set are not from “dry” measurements but rather ambient or “quasi-ambient.” Almost all airborne data falls into this category. Hence the second most important factor in this intercomparison is the assumed hygroscopic growth factor. Because sea-salt particles are generated “wet,” and the relative humidity in the marine boundary layer is always well above efflorescence relative humidity for sea salt, we are only interested in the upper hysteresis curve. For comparison purposes in the results section, we have used the form of *Gerber* [1985] in a static manner for all particles greater than 1  $\mu\text{m}$  in diameter. However, there are differences in the literature worth noting. The first adopted formulations were in a series of papers starting from *Fitzgerald* [1975] and *Hanel* [1976]. This was then modified by *Gerber* [1985] to account for the Kelvin effect. The Gerber formulation has been popular because it gives an empirical solution that is easily applied to models (it is for this reason that we use it in this manuscript).

[79] Variations in hygroscopic growth curves from these formulations, based on inorganic components, exist on the order of  $\pm 10\%$  in size (or 30% in volume). On the basis of the work of *Tang et al.* [1997], small differences in assumed inorganic fractions for fresh seawater do not appear to make any significant difference in the upper part of the sea-salt hysteresis curve. Indeed, with the exception of a slight depression in the efflorescence RH (from 48 to 44%) there is little difference in particle drying compared with simple NaCl. There are, however, small differences in hygroscopicity functional forms in the literature. For example, the curves given in the work of *Tang et al.* [1997] are slightly lower ( $\sim 5\%$ ) than the *Gerber* [1985] parameterization for RH values above 70%. This alone then leads to a 15% baseline uncertainty spread for calculated volume.

[80] Despite reasonable agreement for the theoretical hygroscopic growth of the fresh inorganic sea-salt particles, in reality there are likely to be deviations. For example, open ocean measurements by *Hanel* [1976] give fairly low values ( $d/d_0 = 1.6$  at 80% RH), although this may be due to technique errors. The two most significant deviations are due to chlorine depletion and the presence of organics. Because chlorine is replaced by larger molecules of sulfate or nitrate during aging, the resulting growth curve would be suppressed by up to 10%.

[81] Suppression by organic species is more problematic. *Ming and Russell* [2001] found that for RH values over 75%, hygroscopic growth of fine mode sea-salt particles was suppressed by  $\sim 15\%$  for a 30% organic mass fraction.

Using their same thermodynamic model, *Crahan et al.* [2004] applied filter chemistry from this study and calculated a similar depression for coarse mode particles. Their calculations indicate that for a  $\sim 5 \mu\text{m}$  particle at 85% RH,  $d_{\text{amb}}/d_{\text{dry}}$  ranged from 2.14 to 1.87 to 1.55 for a pure inorganic, 10% organic and 40% organic mass fractions, respectively. This is compared to a value of 2.28 for the *Hanel/Fitzgerald/Gerber* sea-salt parameterization.

[82] Despite the frequent applications of hygroscopic growth curves, and the common assumption that  $d_{80}/d_{\text{dry}}$  is simply 2 for sea salt, in reality hygroscopic growth formulations cannot be treated as trivial, and each individual calculation of hygroscopic growth is very labor intensive. This is particularly true with respect to “column closure” calculation as demonstrated in Figure 8f. A complete study of the hygroscopic properties for the RED study can be found in the work of *Crahan et al.* [2004] and future papers. Our results here support those of *Crahan et al.* [2004] that the Gerber parameterization we used or that of *Tang et al.* [1997] overestimates hygroscopic growth, but it is unclear by how much. The updated chemistry in the inorganic component of the thermodynamic model of *Ming and Russell* [2001] alone suggests a 10% decrease. The organic component of sea salt is much more uncertain. On the basis of *Crahan et al.* [2004], the further inclusion of an organic mass fraction of 10% represented by palmitic acid seems to be appropriate, and would result in an  $\sim 18\%$  total reduction in hygroscopic growth. This would translate into a  $\sim 50\%$  uncertainty range in the calculation of ambient volume from aerosol dry mass, and  $\sim 20\text{--}30\%$  for light scattering. These are significant uncertainties that are typically ignored in sea-salt transport models.

#### 4.3. Vertical Distribution of Sea Salt

[83] One last issue when considering the intercomparison of aircraft and surface instrumentation is the vertical profile of sea salt. Our complete analysis of the vertical distribution of sea salt will be presented in a future manuscript, but, because of its importance in comparing surface to airborne measurements, the state of uncertainty of sea-salt vertical distribution is well worth noting. Under white-capping conditions, and subsequent sea-salt production, a gradient in sea-salt concentrations must form at the surface. We have tried to minimize differences between the near surface and upper level aerosol concentration by choosing days for in-depth comparison where surface production was minimal. In the case of 31 August with 12 m/s wind speeds, we did see some *FLIP*/Twin Otter concentrations ( $\sim 20\%$ ), but this should not significantly influence our primary findings. In their review, *Lewis and Schwartz* [2004] came to a similar conclusion.

#### 4.4. Aerodynamic Particle Sizing

[84] Aerodynamic particle sizers have been reported to have a good response to marine aerosol particles for near-dry particles. For the open ocean, *Quinn et al.* [1996, 1998, 2000] found good closure between their APS 3300 size distributions of dried sampled particles (to 55% RH) with measured light scattering and chemistry. Similarly, the multiple regression results from the PRIDE field campaign show strong mass closure between an APS 3300 and filter measurements [*Maring et al.*, 2003a, 2003b]. It was not



then surprising for the APS data from the RED campaign to also have reasonable mass closure with onboard filter measurements. The relatively good performance of the ground-based APS in this study, as well as the experience of others, suggests that at least in the 1–10  $\mu\text{m}$  diameter range this instrument gives some of the more reliable results (this is of course assuming one employs the corrections of *Armendariz and Leith* [2002] and the APS model 3320 is upgraded to the 3321 to correct for the recirculation problem which results in a spurious  $>10 \mu\text{m}$  peak).

[85] The size distributions measured by *Quinn et al.* [1996, 2000], *Maring et al.* [2003a, 2003b] and this study are very similar (Table 1). APS data collected at 14 m above the surf zone (i.e., shown to be uninfluenced by the surf) in Hawaii by *Clarke et al.* [2003] are also similar. Considering the possible variance in these study's locations and sampled air mass histories, we find all of these studies to be in surprisingly good agreement.

[86] Despite the good performance of the APS on the *FLIP*, the performance of the wing mounted AAPS on the *CIRPAS* Twin Otter does demonstrate that some care must be taken in its employment. Certainly, as in all aircraft or shipboard measurements, inlet issues must be taken into account. Even so, fundamentally, there are several open questions that require addressing in the scientific community. First and foremost, the effects of internal heating by the APS and AAPS on particle size are likely candidates for bias. In our study on *FLIP*, and *Maring et al.* [2003a, 2003b], the aerosol stream was dried to RH values less than 30%. Hence internal heating was not an issue. In the case of *Quinn et al.* [1996], the inlet flow was dried to 55% RH, and hence any further drying would at most make a 30% size correction, to complete efflorescence. In reality, the drying is probably not as great, perhaps on the order of 10–15%. In the case of the AAPS, however, particle sampling is typically at or above 80% RH in the MBL, where the hygroscopicity curve is steepest. We hypothesize that the biases detected in this manuscript are from the convergence of internal heating, drying due to the jet pressure drop, and inlet issues, resulting in a “self sustaining” size distribution in the AAPS.

[87] It is known that hot sheath flow from the pump in particle counters such as the APS and PCASP can cause particle vaporization and even the acceleration of a particle through a jet will cause drying (H. Maring, personal communication, 2004). To mitigate this issue, the AAPS was designed with a considerably longer duct from the pump, in the hope that the sheath flow would equilibrate with the ambient air. However, this may not be enough. The internal temperature of wing-mounted probes can be quite hot. Indeed, as we showed with the PCASP, the surface “undried” size distribution was nearly identical to the “dried” airborne size distribution. If the internal temperature of the AAPS is also high, then the longer sheath air ducting may not be effectual. Unfortunately, the AAPS does not log flow and instrument temperatures, so this hypothesis cannot be tested at this time.

[88] A second (although less likely) possibility is direct radiative heating from the walls. If the surrounding instrument heats the nozzle assembly, then it can directly radiate to the particles. In this mechanism, the temperature of the air around the particles does not need to be heated. Just as the

bulk of the energy at the sea surface goes into the latent heat flux, radiative energy to the particles could evaporate water.

[89] Another issue of interest for the APS is the reaction of particles of variable shape. The calibration of the APS is based on the acceleration rate of spherical particles through a jet. Because particles flow through the APS at super-Stokesian velocities, irregular particles can deviate from the velocity to particle size calibration curves in unpredictable ways. Calibration studies have suggested that the APS under-sizes particles with irregular shapes, such as dust. *Marshall et al.* [1991] suggested the APS could undersize particles by 25% for dynamic shape factor ( $\chi$ ) values of 1.2. For dust,  $\chi$  is often taken as having values of 1.2 to 2. Even so, this ultra-Stokesian uncertainty is rarely taken into account by the scientific community and, even in the PRIDE field study, *Maring et al.* [2003a] managed a fairly good agreement in mass closure without it. This question may still require further research since it affects the study of sea salt in a number of ways. First, while ambient sea-salt particles are hydrated and hence spherical, completely dried sea salt can form complicated structures, though it preferentially forms cubes, which would have a  $\chi$  value of 1.1. However, sea-salt particle measurements have always been done under high vacuum in an electron microscope. Under “dried” conditions in a sampling lines, *Tang et al.* [1997] states that 20% of the mass is still tightly bound water. Under this circumstance it is unclear what the morphological shape is, though it probably tends toward sphericity. The second morphological issue is droplet deformation. What if our previous hypothesis on intrinsic particle drying in the APS is false, and particles are in fact at equilibrium with the outside environment when sampled. It has been hypothesized that in the accelerated jet, liquid drops can deform thus altering their sampling properties in an APS [*Chen et al.*, 1990].

#### 4.5. Cascade Impactor Data

[90] In this study we did not employ an aerosol impactor gravimetric analysis, but it is worth some effort to compare these results to the APSs. In Table 1, we have isolated some of the more frequently referenced sea-salt impactor data sets. On a whole, the reported mass median diameter (MMD) for sea-salt spans from 3 to 8  $\mu\text{m}$ . However, the bulk of the measurements are comparable to the APS data, ranging from 4 to 5  $\mu\text{m}$  at ambient conditions. *Quinn et al.* [1996, 2000] and Savoie (unpublished but with some data in the work of *Reid et al.* [2003]) both showed excellent agreement in estimated size with their individual APS instruments. These findings are also in agreement with the impactor results of *McGovern et al.* [1994] and *Marks* [1990] (it is note worthy that the plots in the work of *Marks* [1990] are for the impactor endpoints, not the midpoints. We have corrected this in our table).

[91] There are two outliers. The largest outlier in this data set is that of *Hoppel et al.* [1989], with reported ambient MMDs ranging from 8 to 10  $\mu\text{m}$ . These data were collected as part of a set of cruises in the Atlantic Ocean in 1983, and are one of the earliest open ocean “local closure” experiments undertaken for size distribution and light scattering. Unlike the more classic cascade impactors listed in Table 1, *Hoppel et al.* [1989] used the Calspan gel replicator system, from which individual particle impacts were analyzed under

a microscope. W. Hoppel (personal communication, 2002) reported that this system was calibrated using FSSP-100 and CSASPs. Hence by inference it was miscalibrated due to the known bias in the FSSP derivatives.

[92] Another outlier is the cascade impactor data of *Howell and Huebert* [1998], here using a MOUDI impactor during the ASTEX experiment in the western Atlantic, as well as at Christmas Island in the central subtropical Pacific. In this case, they found similar particle sea-salt size distributions with MMDs around 7 to 8  $\mu\text{m}$ .

[93] The difference with *Howell and Huebert* [1998] is much more difficult to explain. With a VMD  $\sim 7 \mu\text{m}$ , this is a fairly large impactor outlier. Because all of their size distributions are systematically the same regardless of location (e.g., east Atlantic, or subtropical Pacific), this difference is unlikely to be due to natural variability, and is probably due to some systematic sampling difference between it and the other measurements. This is not to say it is necessarily incorrect. The larger numbers of investigators with VMD values averaging 4–5  $\mu\text{m}$  is not justification alone to throw this measurement out. Indeed, the majority of investigators use optical particle counters, which we know are biased. There is the possibility that the data were impacted by local surf zone, however. *Clarke et al.* [2003] and subsequently released unpublished APS data from this study found that while “background” sea-salt volume distinctions are similar to other APS data sets, samples of surf zone particles have much larger VMDs (7 to 8  $\mu\text{m}$  at ambient conditions) identical to *Howell and Huebert* [1998]. This will be discussed further where we examine the possibility of the null hypothesis.

#### 4.6. Optical Particle Counters

[94] There have been a number of optical particle counter intercomparisons in the literature going back several decades that have shown significant differences between collocated instruments [e.g., *Pinnick and Auvermann*, 1979; *Jensen et al.*, 1983; *Smith*, 1995]. The significance of these differences would be even clearer if these authors plotted the data as volume instead of number. Differences between cloud droplet sizers over the past two decades are also noteworthy [e.g., *Baumgardner*, 1993; *Reid et al.*, 1999; H. Gerber, personal communication, 2003]. All of these studies (plus our own cloud data to be presented in a forthcoming paper) have noted differences between optical particle counters of over an order of magnitude for individual channels.

[95] In this study we demonstrated two principal artifacts in forward scattering probes such as the FSSP, CSASP and CAPS. First, there is a systematic overestimation of particle size due to the inflection in the response curve in the 3–10  $\mu\text{m}$  range. Second, the known reduction in sampling cross section for the first channel certainly extends to the second and probably in part to the third channel as well. This results in an underestimate in the first part of the size spectrum, which in turn results in an overestimation in volume median diameter.

[96] The realization that forward scattering probes have an issue with the inflection point in the response function is by no means new, and we found the topic first discussed in the work of *Pinnick and Auvermann* [1979], *Schacher et al.* [1981], and in more detail by *Baumgardner et al.* [1992].

Similarly, *Collins et al.* [2000] and subsequently *Reid et al.* [2003] for the ACE-2 and PRIDE campaigns, respectively, expended a great deal of effort mitigating the issue. Even so, for the most part the degeneracy issue has been ignored or insufficiently accounted for by the bulk of investigators.

[97] Suggested mitigation methods in the past include: (1) masking the data in the overlap zone and simply extrapolating between the smallest and largest sizes [e.g., *Schacher et al.*, 1981], (2) grouping bins together into three or four independent “super bins,” with one covering the entire degeneracy region [e.g., *Collins et al.*, 2000], or (3) when possible, proceed strictly in “voltage space” from the raw instrument data and try to directly derive related parameters such as light scattering [e.g., *Reid et al.*, 2003]. None of these methods are satisfying, and all are difficult to implement into parameterizations suitable for a model. The crux of the problem is that it is difficult to derive a consistent size distribution for all three moments simultaneously (number, surface area, and volume). Because the number distribution falls off so precipitously in the coarse mode, the number concentration in any bin is heavily weighted to the lower bin boundary. Any slight deviation in the number distribution will be amplified in the surface ( $d_p^2$ ) and volume distributions ( $d_p^3$ ). In regard to method a, examination of the response curve in the work of *Collins et al.* [2000] or *Reid et al.* [2003] with the above analysis shows that, while there is a significant degeneracy zone centered around 5  $\mu\text{m}$ , degeneracy does occur to some extent throughout the entire curve. So, selectively removing bins will not entirely solve the problem, but it can produce a more reasonable answer in size, but perhaps not in amplitude [e.g., *Fairall et al.*, 1983]. This also makes the distribution curve highly subjective. By rebinning (method b), we at least force a more uncertain interpretation of the VMD, but we are still left with the issue of missizing across the “super-bin” boundaries. Further, the bin starting point will control the number concentration, and wider bins will have higher mean geometric diameters, thus biasing total surface and volume distributions further. Hence the total volume will still be overestimated by a variable amount (i.e., dependent on the true size distribution). Finally, (method c) we can interpret based on voltage space. However, while this is more mathematically pure, and allows the data to be used in “closure-like” studies, it cannot derive the fundamental tangible parameters needed by modelers.

[98] Ultimately, the degeneracy issue explained almost all of the bias in OPCs found by *Reid et al.* [2003] for dust particles. However, in the case of sea salt (Table 1) we find considerably more scatter in OPC data, with some reported values near or even below the values given by aerodynamic methods. However, with the exception of two cases (discussed later) all upper end OPC reports are from single gain stage instruments. The “more correct” reports from OPCs are from multi gain stage instruments [e.g., *Gerber*, 1985; *Horvath et al.*, 1990]. This effect is in part due to the offsetting error of the undercounting in the first two to three channels in each gain stage and subjectivity in apply the curve fit (as demonstrated earlier in this manuscript). Particle underestimates in early channels can compensate for particle over-sizing in others. The result depends on how the various gain stages are weighted. By having more gain stages, the size distribution can become more uncertain.

[99] In addition to the biases described above, we must also consider other instrument specific biases. Even identical instruments, placed side by side and given identical calibrations, have shown significant variance [e.g., *Jensen et al.*, 1983]. The fact that instruments are calibrated for size, but not always sampling volume, can account for part of this variance. Noise in the system, dirty prisms and simple electronic variability (in original PMS versions) can also have significant, consistent impacts in all OPCs. Early closed celled systems such as the ASASP suffer from much greater inlet loss and drying issues.

[100] In all cases, the issue of curve fitting is extremely important. A least squares curve fit around a number distribution would not likely be a good fit for higher moments such as area and volume. Indeed, a brief examination of the raw data from *van Eijk and de Leeuw* [1992] provided by the authors showed a 4 fold discrepancy between the lognormal number distribution fit-derived VMD and that actually in the raw data (Table 1). A combination of these factors explain the very low VMD in the Navy Aerosol Model (VMD = 2.1  $\mu\text{m}$ ) reported by *Gathman* [1982, 1983] and is the topic of a future manuscript.

[101] Ultimately, the findings of this manuscript demonstrate that the data from all FSSP like instruments should be treated with extreme caution. In well-calibrated and maintained instruments, the biases discussed above converge to result in an overestimation of sea-salt size and volume. Indeed, the transfer function between size and voltage dictates this should be the case. Our own Monte Carlo simulations show that regardless of the input size distribution for VMDs between 2 and 10  $\mu\text{m}$ , the output VMD is the same,  $\sim 8\text{--}10\ \mu\text{m}$ . Consequently, much of the community field data on sea salt from PMS style OPC systems, as presented, should be treated with a great deal of skepticism. This does not, however, make the data useless, nor have implications for instruments not studied in this particular manuscript. Indeed, the simple fact FSSP and CSASP are so prominent in the literature, and that the FSSP and CAPS are the only open celled wing mounted probes should make the scientific community take them seriously. All of the recommendations on data utility by *Reid et al.* [2003] for dust hold for sea salt. If placed in the correct context, these instruments can be quite valuable for calculating particle gradients, optical properties and, as will be a topic of a future paper, sea-salt particle fluxes.

#### 4.7. Sun/Sky Inversions

[102] AERONET-based inversions using the Dubovik retrieval are becoming the mainstay of global model validation. However, the retrievals are for the most part unvalidated. While we were not in a position to perform a complete validation program during the mission, our findings do weigh in on the issue. As the VMD and geometric standard deviation did not appreciably vary at Lanai during the study, and we do not have vertical profiles over that particular site, we cannot establish distribution sensitivity. However, the dry size distributions from APS data on the *FLIP* were also fairly static in shape, and only altered in magnitude, similar to the Lanai site. As the retrievals were constrained by optical depth, we know that the inverted size distributions do at least reproduce the basic optical proper-

ties of the atmosphere. We can also say that the shape of the size distribution curves at the Lanai site were closer to the APS “secondary standard” than any of the wing mounted probes on the Twin Otter, or the other optical particle counters on the *FLIP*. Indeed, we find that our model of column-integrated size overestimates AOT by roughly 30–45% (to be discussed in a subsequent paper). The low optical depth of the marine environment, coupled with the sampling, sizing and hygroscopicity uncertainties in the in situ measurements, converge to create a near impossible situation with regard to “column closure.” However, under the limited constraints of this study, it does appear that when *Dubovik and King* [2000] inversion is applied to properly quality assured data, the results are consistent with what we know of the marine environment. This does not mean, however, that we consider the algorithm fully evaluated. For example, the volume distribution seems fairly wide in comparison to our measurements. Because of the factor 5 difference between the mass extinction efficiency of the fine and coarse modes, slight perturbations in the fine mode size and concentration can have a significant effect on the magnitude of the volume distribution (e.g., to retain the AOT constraint). Even so, the retrievals do appear to be more consistent with the true marine environment than most other in situ column-integrated measurements in the field.

#### 5. Implications and Recommendations

[103] The measurement and modeling of sea-salt influences a number of fields, and it is worth briefly examining the impact of our findings, and how they propagate. The primary implication of this study is that the bulk of the FSSP/CSAPS/CAPS and likely ASASP-based data used in the past needs to be eyed with suspicion and probably excluded in any consensus. Investigators that have blindly averaged all data into a uniform model risk significant bias. With the exception of the *Hoppel et al.* [1989] and *Howell and Huebert* [1998] reports discussed above, all of the aerodynamic methods converge further to a coarse mode VMD in the 4 to 5  $\mu\text{m}$  range. Some OPC data does exist showing VMD in the 5 to 6  $\mu\text{m}$  range. At this point, however, the issue becomes less resolvable. It can be argued that almost all OPC systems must deal with the degeneracy/index of refraction issues to some extent, suggesting that these would likely be overestimates in VMD, even if only slightly. Conversely, all impactor data must cope with sampling and inlet issues, which can certainly be significant in marine conditions when high winds are not uncommon. Thus these systems, under some conditions, would likely underestimate the VMD. On the basis of all of these considerations we can only recommend an 80% RH mean value between these groups or  $5 + 0.5\text{--}1\ \mu\text{m}$  VMD (note asymmetric error) and a geometric standard deviation of  $\sim 1.8\text{--}2$ . This is significantly different from a number of highly used marine models, including the Navy Aerosol Model (NAM) of *Gathman* [1982, 1983], *Porter and Clarke* [1997] and the recently proposed model of *Lewis and Schwartz* [2004].

[104] Most fundamental to modeling endeavors is the application of source and sink functions. As discussed in the introduction, reported and applied source functions vary by orders of magnitude. What fraction of this is due to the



size distributions themselves? Almost all flux measurements in the literature for open ocean conditions are based on OPC systems, and four [Fairall *et al.*, 1983; Monahan *et al.*, 1986; Smith *et al.*, 1993; Reid *et al.*, 2001] all used FSSP-like systems in particular. Consequently, all subsequent studies that employed these fluxes [e.g., Andreas, 1998; Lewis and Schwartz, 2004] should also be re-examined. In the Smith *et al.* [1993] and Reid *et al.* [2001] method, FSSP data were used without any smoothing function, and clearly the anomalous VMD peak at  $\sim 8 \mu\text{m}$  exists. For Monahan *et al.* [1986] and Fairall *et al.* [1983], the smoothing function of Schacher *et al.* [1981] was applied, and hence these are based on a particle VMD of  $\sim 4\text{--}5 \mu\text{m}$ . This does not imply that they are necessarily correct in magnitude, only that the relative size bias has been mitigated. Ironically, the Monahan and Fairall schemes upper and lower bound the more commonly used flux algorithms, and the Reid findings match those of Monahan. This suggests that while coarse mode particle flux algorithms are certainly biased by the almost universal use of FSSP like systems, there are much bigger issues still at large.

[105] A second geophysical quantity of interest is the sea-salt mass concentration itself. The above reports influence the aerosol science community greatly, in that all sea-salt effects are ultimately based on the actual particle concentration, either through direct forward modeling or through the validation of global climate or chemical transport models. Like the very large variance in flux algorithms, the thorough compilation of published sea-salt concentration data and parameterizations by Lewis and Schwartz [2004] shows differences of an order of magnitude. However, unlike the fluxes, this variance cannot be easily tracked back to specific methods. Certainly, the use of FSSP like systems will lead to significant overestimates in sea-salt mass concentration. Closed cell OPC systems also have the possibility to underestimate particle concentration (through uncorrected drying, inlet issues etc). However, filter and impactor-based methods also show tremendous variance. A large fraction of the chemistry and gravimetry results are undoubtedly due to natural variability. Inlet cut point, and other sampling issues are probably also significant. In a retrospective analysis, it is difficult to evaluate the quality of some of these data. However, our results clearly show the importance of having overlapping/over-sampling methods. Certainly the use of an APS, coupled with filters and impactors, is the most straightforward way to ensure consistency in the data set. While such deployments are not uncommon, large consistent data sets suitable for model validation are. In our searches, only the data from long cruises presented in the work of Quinn *et al.* [1998, 2000] met these requirements. This does not imply that the countless other data sets are without merit, but rather that they (or any other data sets for that matter) should not be used blindly and certainly not “bin averaged.” Trends in such data sets are likely real, but extreme caution must be exercised in using absolute values. This is particularly true with regard to model validation, where we know significant biases exist in both the source term and the validation data.

[106] The errors in concentration and size subsequently feed back into other areas such as radiation and chemistry. Aside from the propagation of biases in simple concentration, errors in coarse mode size parameterizations can cause

confusion in the field, particularly with regard to the application of field data to modeling studies. A discussion of the impact of changes in coarse mode particle size on the mass scattering efficiency is presented in the work of Reid *et al.* [2003] (while this is for dust, the same trends apply for sea salt). Simply put, for coarse mode particles  $Q_{\text{ext}}$  is relatively constant at  $\sim 2$ , the cross sectional area goes as  $d^2$  with size, and the volume as  $d^3$ , and hence the mass scattering efficiency goes inversely with diameter. If we look at the range of VMDs reported in the literature we have more than a factor of 5 in potential variance and any modeled error in size will linearly affect any radiation effect.

[107] The crux of the sea-salt size-radiation problem, however, is that a single lognormal volume or number distribution does not adequately represent all the micro-physical aspects of coarse mode sea salt. Recall, there is the possibility that jet drops control the volume distribution, whereas the number distribution is more influenced by film production. As calculated by Quinn *et al.* [1996], the bulk of scattering by sea salt in the visible is not around the volume median diameter ( $\sim 3\text{--}6 \mu\text{m}$ ), but rather on the forward tail, where the scattering kernel is largest ( $\sim 0.5\text{--}2 \mu\text{m}$ ). Hence while we parameterize particles based on their volume distribution, and can perform sensitivity cases based on the perturbation of the volume median diameter and standard deviation, in actuality it is the beginning of the distribution that often controls light scattering (e.g., “tail wagging the dog”). Similar arguments can be made with the number distribution. Surface area distributions are cumbersome and not directly measurable.

[108] Particles that control light scattering are often parameterized using instruments such as the PCASP, or better, differential mobility analyzers. Hence in a “closure” test, it is possible to achieve closure with light extinction or optical depth, even if the coarse VMD or even total mass concentration is biased. However, unfortunately, in terms of large data sets suitable for GCM or CTM model applications, validation is almost entirely done with volume or mass measurements. Again, light scattering, AOT, and size must be measured simultaneously in order to truly validate these simulations.

[109] If anything, this manuscript clearly shows that the scientific community cannot continue using the same methodologies as employed in the past. At the present time, there has not been demonstrated a sampling system that can measure the ambient sea-salt particle size distribution from an aircraft. This is a problem with broad scientific implications. In the context of this manuscript, for example, we cannot validate the AERONET inversions. One of the largest areas of concern is that one of the largest uncertainties in the measurement of indirect forcing is likely in the measurement methods themselves. This is particularly true for very high humidity conditions, such as those present at cloud base. None of the instruments used in this manuscript can “observe” the transition of an “aerosol particle” into a cloud droplet. This is particularly important if we are to assess the hypothesis modeled in the work of Feingold *et al.* [1999], that coarse mode sea salt can counteract indirect forcing by anthropogenic particles.

[110] Fortunately, the scientific community is reacting to the situation. In just the past two years, recognition of these issues has led to the development of white light-based

OPCs that are far less sensitive to degeneracy issues (e.g., the commercial systems by Palas Inc., or the LED systems at the NOAA Aeronomy Laboratory; Wollney *et al.* [2004]). Although these are closed cell systems, they do show great promise in the marine environment and should be considered a high priority for the community. Open celled Phase Doppler Velocimeters are also becoming commercially available (e.g., TSI Inc). While in their infancy, these also hold great promise.

[111] This manuscript also demonstrates the necessity of gravimetric analysis and proper ion chromatography in marine aerosol studies. None of the OPC studies that we have listed as outliers in this study had any form of gravimetry or chemistry presented in their manuscripts. Despite the difficulty, expense and sometimes tedious nature of this sort of measurements, it is absolutely necessary. On aircraft, the labor involved in, say, the total aerosol sampler [Blomquist *et al.*, 2001] is a necessity for marine coarse mode studies.

## 6. Final Assessment and Conclusions

[112] In this manuscript, we intercompared commonly used instruments for sizing coarse mode sea-salt size distributions, hypothesized causes for differences, and discussed impacts. Our findings can be summarized as follows:

[113] 1. Using both the R/P *FLIP* and the CIRPAS Twin Otter, we intercompared the results of ground and airborne Aerodynamic Particle Sizers (APS), Forward Scattering Spectrometer Probes (FSSP), Classical Scattering Aerosol Spectrometer Probe, the airborne Complete Aerosol Probe (CAPS) forward and backscattering components, and chemistry and gravimetry. We also examined column-integrated sea-salt inversions from local AERONET sites. The purpose of this comparison was to try and explain the extreme variance in the peer-reviewed literature regarding coarse mode sea-salt size distributions.

[114] 2. This comparison took place for a two-week period 11 km offshore on the windward side of Oahu, where back trajectories out to 10 days never reached the mainland. These should be considered open ocean conditions. Two large sea-salt events (one local, one transported in) modulated the bulk of the sea-salt concentrations in the  $\sim$ PM<sub>10</sub> range between 3 to 12  $\mu\text{g m}^{-3}$ . Midvisible optical depths ranged from 0.06 to 0.11.

[115] 3. APS data collected on the *FLIP* matched gravimetry better than any other method. The derived size distribution also compared well with other reports in the literature. The airborne APS, wing-mounted on the Twin Otter, however, shows poorer and sometimes unphysical performance. This is particularly true at higher humidity such as at the top of the MBL. We hypothesize that it may suffer from inlet losses as well as internal particle drying. As it is prone to drying in the sampling lines, we recommend that the APS samples be dried to RH values less than 35%. While this comparison is better than any of the other methods, significant issues still remain with the APS and we do not consider the instrument fully validated.

[116] 4. The ground-based FSSP and CSASP and airborne CAPS probes, open celled instruments sampling ambient particles, displayed significant particle over sizing, similar to previous reports for dust. Derived dry mass concentrations

from these instruments were high by over a factor of two to three compared to gravimetric measurements. This is a result of two principal mechanisms: Uncorrectable degeneracy in the response curve and undercounting in the first 2 to 3 bins. In the case of the airborne FSSP running at the lowest gain settings (i.e., set for cloud observations), this prevents any quantitative use of the data for coarse mode aerosol studies.

[117] 5. The multigain stage CSASP correlated extremely well with the *FLIP* FSSP, although the magnitude in volume is highly subjective based on curve fitting parameters. The CSASP gain stage 1 and the *FLIP* FSSP were almost identical.

[118] 6. The instrument biases listed above, as well as inlet, curve fit, and reporting biases, explain the bulk of the variability in reports of coarse mode particle size. We find that a VMD on the order of  $5 + 0.5/-1 \mu\text{m}$  is a likely mean candidate for sea salt at an 80% RH. The finding of this study combined with others that employ aerodynamic or impaction systems suggest that the size parameters for coarse mode sea salt may be surprisingly invariant under most conditions.

[119] 7. Our findings also point to the importance of proper hygroscopicity parameterizations for sea salt. Our findings are consistent with the idea that the standard hygroscopicity curves overestimate hygroscopic growth for sea salt, and that the impacts of organics need to be taken into account [Ming and Russell, 2001; Crahan *et al.*, 2004].

[120] 8. Most seriously, our findings also suggest that currently there has not been demonstrated a real time particle sizing method that can quantitatively measure the ambient size distribution of coarse mode sea-salt particles from an aircraft. In particular, the interpretation of the vertical distribution of sea-salt size distributions in marine boundary layers is complicated. This does not imply that existing instruments are without value. However, these instruments are now being applied to problems for which they were not originally engineered and the needs of the scientific community have outstripped their applicability. It then follows that perhaps the largest uncertainty on marine aerosol studies of radiation and aerosol particle/cloud interaction is the instrumentation itself.

[121] 9. The Dubovik and King retrievals from local AERONET sites were also evaluated. While local effects prevented a direct validation of the product, derived coarse mode size distributions appear to be physically reasonable. Derived column-integrated coarse mode volume matched gravimetry surprisingly well. However, given the uncertainties in particle measurement from aircraft, it is difficult to validate the inversion fully.

[122] 10. Our findings also show a consistent propagation of particle sizing error through the literature from a variety of sources regarding geochemical cycles, fluxes, chemistry, and radiation. In many cases, the instrument errors would not necessarily be decipherable in radiation internal closure studies, but would rather manifest themselves later when derived parameterizations were incorporated into models. Regardless, the bulk of empirical sea-salt concentration and flux parameterizations are more suspect than ever.

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