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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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 μm for almost every method. Can this variability be reconciled?

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 ~PM10 range between 3 to 12 μg 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 μm 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.