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Atmospheric Measurement Techniques Discussions

Interactive comment on "A Marine Aerosol Reference Tank system as a breaking wave analogue" *by* M. D. Stokes et al.

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Can we explain the apparent bimodal character of the MART SSA size distribution with a dip at 700nm (Figure 5)...

This is correct and text has been added to the revised manuscript to further clarify this observation. To illustrate the SSA size distribution over such an extended size range requires an amalgamation of particle sizing techniques from 2 different instruments. Because the particle measurements from the SMPS tends to undercount particles at the high end of the distribution due to the cut-off from the particle impactor, while the APS can undercount particles at the low end due to poor scattering efficiency of the smallest particles. As a result, particle bins in the overlapping size region of the two

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methods were subsequently removed, excluding the largest and smallest bins of the SMPS and APS, respectively (Figure 5). Because of this, caution is required in any detailed analysis of the overlap region between the two measurement methods, at or around 700 nm (Figure 5). And the slight dip in the distribution is most likely an artifact of the amalgamation. As indicated by the Reviewer, this is also noted in the lab study reported by Prather et al. (now 2013), which is not yet added to the reference list (it is in secondary review, but hope to be listed as "in press" very soon). The full Prather reference will be: Prather et al. Bringing the ocean into the laboratory: Impacts of chemical complexity of sea spray aerosol on climate relevant properties. Proceedings of the National Academy of Sciences, 2013.

The use of the Teflon tank by the Fuentes study to minimize contamination. Can we insure that the MART system is free from organic contaminants in the generated SSA's from both the plexiglass tank and centrifugal pump?

As noted in the text, we attempt to minimize contamination in the MART system by carefully cleaning the tank, pump, tubing, plumbing etc. with a standard protocol involving alcohol rinses, bleach (if culturing live material), elbow grease and lengthy flushing with de-ionized fresh water. Because of the plexiglass tank more caustic chemical cleaning treatments (possible with an all Teflon system) eliminating all background organic compounds is not possible. However, as a baseline we continue cleaning the MART system until the surface tension of the water within, after the pump is activated, is measured as 72 dyne/cm and close to that of 'pure' seawater. Scanning transmission X-ray microscopy of particles generated from an artificial sea salt matrix within MART suggests minimal single particle carbon content. Future work is aimed at quantifying exact organic carbon concentrations that might be sourced from any impurities in the MART system. And, as noted in the text, ideally, the MART system could be fabricated entirely out of other materials (like Teflon, stainless steel, glass) to minimize contamination and facilitate more vigorous chemical cleaning.

What are the relative role of jet and film drops in the aerosol production? Does the

AMTD

5, C4090–C4094, 2013

Hinze scale define a boundary above which bubbles burst to form particles, and below is stabilized by surface tension? Does it apply to the surface interface as well as in the bubble plume? Does the tank mimic interface conditions as well as those in the plume? See page 8705.

As is noted by the Reviewer, there is not an extensive literature on this topic. The large bubbles are important because the film-drop and jet drop SSA production mechanisms are scale-dependent. It is generally accepted that film drops are produced by bubbles larger than roughly 1 mm radius (e.g. see Fig. 30 in Lewis and Schwartz, p. 206), whereas jet drops are produced in quantities greater than 1 per bubble by bubbles less than 1.5 mm radius (see Fig. 26 in L&S, p. 191). Physical production mechanisms that do not reproduce bubbles at the large end of the spectrum will therefore preferentially enhance jet drop over film drop production in comparison with the ratio expected from breaking waves. Exactly to what extent the SSA production processes will be biased is complicated by the fact that bubbles floating on the surface have a different and surfactant-dependant shape from that of rising bubbles (M. Nicolson, Proc. Cambridge Philos. Soc. 45, 288, 1949) and, once on the surface, foam coarsens through a process of bubble coalesce which creates films of increasing scale (A. Saint-Jalmes, "Physical chemistry in foam drainage and coarsening", Soft Matter, 2 836-849, 2006). This complicated topic lies beyond the scope of the present article. We do feel that facilities like MART will help us explore these issues. The revised manuscript has had text added to clarify these points and the additional references added to the cited literature.

Can MART be used to study foam decay as well as pseudo-steady state foams which is important for SSA generation (page 8713)? Yes, the MART system can be used to study foam evolution and is currently being used in that fashion in new studies. Text has been added to the revised manuscript with further details. For example, by manually activating the plunging sheet to produce isolated foam patches or changing the TR53122 timing relay to one enabling longer delay times between plunging, the

AMTD

5, C4090–C4094, 2013

properties of surface foam patches and SSA production as they evolve over time can be examined.

Line 11, Page 8711. “The methodology for determining the deposition in the tank using the UHSAS does highlight the difficulty with, in particular, the determination of the formation rate of larger particles,. . .” but the statements on Line 11 should be further clarified concerning empirical corrections.

As indicated by the Reviewer this phrasing has been clarified in the revised manuscript. Because of the difficulty in using particle lifetime with respect to deposition which can be dependent on the carrier gas flow rate, a bias in the measured size distribution is expected for particles where τDEP is significantly smaller than τMIX (e.g., dp > 2 μm; τDEP = 1.5 min and τMIX = 90 min for 1 slpm carrier gas flow), highlighting the importance of using size dependent, empirical corrections when determining the size distribution of nascent SSA.

Additional comparison to Fuentes and Sellegri et al. and scaled plunging jet mechanisms (see also Reviewer 1). Additional text has been added to the revised manuscript that includes the jet, “weir” of Sellegri et al..

line 6, p8709 - is it really a 0.058 cm impactor (should it be 580 nm)? The impactor has been confirmed at 508 μm, and corrected in the revised manuscript text.

With respect to particle drying, How was RH measured? What was the residence time in the drier? With an experimentally determined effective density of 1.8 g/cc for the solid particles is this a meaningful shape factor for the dry crystalline NaCl? Are the particles really dry?

Relative humidity was not measured continuously at the exit of the particle drier in this specific experiment. All future measurements made using MART will have an integrated RH sensor following the inline particle driers. Nonetheless, the desiccant used in these experiments was baked at 130 C for over 3 hours prior to this experiment.

AMTD

5, C4090–C4094, 2013

Prior characterization of this specific inline drier has shown the RH at the exit of the drier to be less than 15% for sea-spray aerosol generated with MART. The residence time is estimated to be 3.3 s, based on the volumetric flow rate and drier internal volume. Analysis of the sea-spray particles using Atomic Force Microscopy (AFM) at this RH indicate that the sampled sea-spray particles are not pure NaCl crystals, due to MgCl2 hydrates that retain water.

lines 8-10, p8709 The sentence describing flow rate and flow ratio controls is incomplete. This sentence has been clarified in the revised manuscript. As noted by the Reviewer, the range of particle sizes which can be analyzed and the transfer function of the DMA is dependent on the voltage applied to the central rod and the aerosol and sheath flow rates, which were set at 0.4 and 4.0 L min-1, respectively.

Abstract: line 7 The abstract sentence has been clarified in the revised manuscript and it is now explicitly stated that the water flow is monitored.

Title. The title as is might be suggestive that the MART system is to reproduce all aspects of breaking waves. At the suggestion of the Reviewer, the title has been expanded to indicate that the analogue referred to is for the purpose of foam and SSA generation and not to address all wave dynamics. “A Marine Aerosol Reference Tank system as a breaking wave analogue for the production of foam and sea spray aerosols.”

Interactive comment on Atmos. Meas. Tech. Discuss., 5, 8701, 2012.