Atmos. Meas. Tech. Discuss., 5, C4039–C4044, 2013
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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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Received and published: 8 March 2013

The paper is timely. There have been several efforts over recent years to use bubbletanks to mimic the processes leading to primary marine aerosol formation and the aspiration to develop a reference tank is well-justified. The authors should be commended for the experimental design. The tank appears very well thought through, with a comprehensive array of control and monitoring that should enable a wide range of conditions to be reliably and repeatably investigated. Of particular interest is the intermittency of the flow and replication of the wave crest by the plunging sheet arrangement. The system characterisation methodology appears similarly well-designed, with convincing demonstration of representativeness of bubbles and, broadly, of aerosol. I

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have a few questions about the aerosol measurements at the end of this review, but these are relatively technical and require minor attention. This study addresses many of the problems encountered in previous studies and the manuscript should be published once the authors have addressed the technical and minor queries plus one or two slightly more substantial further points below.

The whole way through the manuscript, there is a quite appropriate focus on the ability of the tank to generate SSA. However, the tank design for this purpose relies on the a priori assumption that the determinant processes for SSA production will be those controlled in the tank setup. An interesting phenomenon evident in figure 5 is the production in the tank of a bimodal aerosol distribution with a peak at around 210 or 220 nm and a dip at 700 nm diameter. It is unclear whether such bimodality indicates a duality of processes in the tank that is not occurring in a breaking wave. In particular, the dip at 700 nm is not evident in the breaking wave nor in ambient distributions. Also, such a large modal size is in contradiction to previously reported ambient and bubbletank spectra. The authors should comment on this beyond comparison with lab studies reported in the Prather et al., 2012 study that is not found in the reference list.

The authors correctly identify that the Fuentes study would have benefitted from a larger tank and are completely balanced in the discussion of this study (though omit consideration of the Froude number scaling employed to justify the tank geometry). However, one of the reasons for the relatively small tank was the requirement for an inert hydrophobic internal surface material, satisfied by the use of solid Teflon sheet sealed at the joints solely by compression with no adhesive or sealant. This was necessitated by the use of micromolar concentrations of organic material to replicate seawater concentrations. Using plastic or even highly cleaned glass, it was found extremely difficult to generate a background aerosol sufficiently clean to remain free of organic contaminants in blank experiments using NaCl solution. The authors should therefore be commended on their ability to clean the tank to attain a surface tension value of 72 dyne/cm. This is pretty convincing evidence for a clean tank, but have the

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authors established that there are no contaminating components in the aerosol particles? Furthermore, only by using the rigorously cleaned Teflon tank was confidence and reproducibility achieved in the organic content of the aerosol for real seawater or proxy algal exudate experiments. The authors should comment on how confidence in such studies of the effects of organic compounds on SSA formation, composition and properties can be achieved in their Plexiglass tank.

It should be noted that previous plunging jet arrangements, where they use peristaltic pumps, also confer intermittency by virtue of the pump motion. Whilst it is more by luck than judgement, this also avoids the interference with a rising bubble plume that would occur under continuous flow conditions. One advantage of a peristaltic pump (when the Teflon pipe is changed sufficiently frequently) is the lack of organic contamination. How is organic contamination in the centrifugal pumps powering the MART system avoided?

A distinct advantage of the current study is the measurement across the entire bubble distribution, out to larger sizes. It seems clear that a system that more closely replicates bubbles across a broader range will be a more representative system. One thing that I am unclear about is the relative contribution of film and jet mode aerosol from the various sizes across the bubble spectrum. I am aware of some studies claiming that most film mode particles come from bubbles > about 2mm diameter and that the jet mode from smaller bubbles (< 10 microns) can be mistakenly identified as film mode. Is the inference within the current study (e.g line 18-19 page 8705) that the Hinze scale defines the boundary above which bubbles burst to form particles, below this being stabilised by surface tension or is there still a significant contribution from the more numerous smaller bubbles. The sentence could be rephrased to clarify this point (and I'd be very pleased to see a few references to any literature unambiguously attributing different modes of particle formation to different parts of the bubble distribution in seawater if any such literature exists). I am no expert in bubble bursting dynamics, but it would be good to know whether the Hinze scale defining the stabilising boundary in

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the rising bubble plume pertains when the bubble reaches the surface and is no longer spherical. Has this been studied and is there a different diameter at which a bubble is stable on the surface than in the water column (the pressure gradient arising from surface tension forces being different when air is on both sides of the film and the shear forces driving turbulence at the water-air interface being different)? I'm not asking for a comprehensive review of the literature here, but think it would be informative to know whether the tank acts as an analogue in the respect that it mimics conditions at the interface as well as in the plume (other than by stating that the tank generates a foam patch).

The discussion of some of the questions of importance to aerosol generation from bubble-bursting on p 8713 (e.g. foam decay as opposed to pseudo-steady state foams) is important. It should be discussed whether the MART system can be used for such studies.

In the conclusion section, lines 18-25 on p8714 outline a recommendation to reproduce bubble plume characteristics to be representative of the ocean. I would probably consider the main criterion for appropriateness as an analogue for SSA generation to be the match to the ambient marine primary aerosol distribution rather than the bubble distribution. If the latter can be matched at the same time, then so much the better. I do generally agree with the authors that an intermittent plunging sheet is a good method of generating SSA, but I am in full agreement with referee 1 that a direct compariosn with an appropriately scaled plunging jet system should be made. Only after the evaluation of each of these systems against ambient wave breaking aerosol distributions would I recommend that any reference system should be universally adopted.

Technical queries:

line 6, p8709 - is it really a 0.058 cm impactor (that is 580 microns, should it not be 580 nm)?

lines 8-10, p8709 - obviously the voltage on the central rod controls the DMA sizes for

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a given flowrate. The flowrate and flow ratio controls the transfer function width and absolute value of the size range for a given voltage range. This sentence should be corrected.

It is stated that the particles are dried. How was the RH measured? It is surprising that the modal diameter is so large (compared with previously reported bubbletank and ambient measurements). This could be explained if the particles still had some associated water, and had not been completely dried out. What was the residence time in the drier? Has a calculation been done to establish that the particles had sufficient time to equilibrate to the ambient RH in the drier?

The dry density of solid NaCl is 2.16 g/cc. It is stated that the effective density is experimentally determined at 1.8 g/cc. The authors should comment on whether this leads to a physically meaningful shape factor for dry crystalline NaCl and therefore whether they are confident that the particles they are measuring are really dry.

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 (and hence determination of supermicron SSA fluxes). On line 11, p8711 - it is stated "... highlighting the importance of running proper aerosol dynamic simulations when determining size distributions". I am unsure of the sort of aerosol dynamics simulations to which the authors are referring. Without a polydisperse aerosol dynamics model coupled to CFD, it is not possible to simulate the processes alluded to. I'm sure / would hope that the authors are not recommending this. Unless they are, the statement should be rephrased to something like "such empirical corrections should be employed when determining the actual distributions present at the point of formation".

Minor points:

Abstract: line 7 - what is measured volumetrically, the amount of water?! It is clear from the body text of the paper, but the abstract should be self-contained.

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Title: it is not clear whether both the intention and achievements of the reference tank were to faithfully reproduce all aspects of breaking waves. A suggestion would be to qualify the purposes of the analogue in the title (i.e. for the purpose of SSA analogue generation).

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