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Atmospheric Measurement Techniques Discussions

Interactive comment on "Laboratory-generated primary marine aerosol via bubble-bursting and atomization" *by* E. Fuentes et al.

Anonymous Referee #1

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General Comments:

The subject matter addressed in this manuscript is important and appropriate for publication in AMT and some of the results, particularly the measurements of size-resolve hygroscopicity, are quite interesting. However, based on the specific comments itemized below, I am do not believe that reported data generated using this experimental design can be reliably interpreted to assess the "validity" of different approaches for artificially generating marine aerosols. Major concerns include the following:

- The shallow tank, short bubble path length (few cm), and limited time for bubble plume evolution are not representative of conditions in the surface ocean in which plumes typically extend to 1 or more m depth.

- The production experiments were conducted using artificial seawater devoid of organics and thus ignored the potentially significant influences of organics on bubble plume dynamics and aerosol production.

- The rationale for selecting the data subsets that were compared is often unclear and does not, in my view, provide an objective context for evaluating behavior.

- Several underlying assumptions employed in data interpretation are not stated explicitly or justified and some are not valid.

In addition, a recent paper by Houtin, Nilsson, et al. [2009, JGR, in press] compares size-resolved production fluxes of artificially generated aerosols reported in the literature (all except Houtin et al are cited in the subject manuscript) using a number of different approaches. Aerosols produced by bubbles from a jet of natural seawater [Houtin et al] and by pushing air through sintered glass in natural seawater [Keene et al., 2007] yielded the best agreement whereas those involving aerosols produced in artificial seawater by both falling water and other methods of aeration diverged to a greater degree. These results suggest that, for the generation of representative marine aerosol, the nature of the aqueous medium may be a more important than the bubble production mechanism. In contrast to results reported by Houtine et al., Fuentes et al. report relatively large differences in aerosol size distributions generated by a water jet versus sintered glass using artificial seawater devoid of organics. Available evidence cited herein supports my suspicion that the differences reported by Fuentes et al. may be specific to the experimental design and are not broadly applicable.

In additional to addressing substantive issues, the specific comments below offer a number of suggestions for clarifying the presentation. In this regard, it would also be helpful to consolidate all relevant experimental conditions used to generate the reported data into a single table.

Elements of the manuscript may be suitable for publication after major revision to address concerns raised herein but I cannot recommend publication in this or similar

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form.

Specific Comments:

Page 2282, starting on line 2. The abstract requires revision based on the points raised below.

Page 2283, lines 3-5. While the production of primary marine aerosol is the dominant natural source for atmospheric aerosols globally, available evidence indicates that the corresponding burden of non-water mass is similar to that of naturally produced mineral aerosol [e.g., Andreae and Rosenfeld, 2008, Earth-Sci Rev.]. In the literature, "loading" is often used in the context of a burden rather than a flux. To minimize the potential for confusion, this distinction should be clarified in the text

Page 2283, line 13. The upper size limit for "sea-salt droplets" produced by breaking waves at the ocean surface is typically much greater than 10-μm diameter [e.g., see several of the cited papers as well as the comprehensive review by Lewis and Schwartz, 2004, Geophysical monograph 152, AGU]. The text should be corrected.

Page 2284, lines 6-9. Here and elsewhere in the text, use of the term "sea-salt particles" is ambiguous. Certainly, inorganic sea-salt species are major constituents of aerosols in marine regions. However, some individual particles in surface marine air originate from continental sources and/or nucleation pathways and virtually all contain both inorganic and organic constituents that originate from secondary pathways involving both marine and terrestrial precursors. A general term such as "aerosols in near-surface marine air" would be a more appropriate descriptor.

In addition, this section implies that the composition of aerosols in marine air at a given location can be interpreted directly in terms of biological processes in the underlying ocean at that location. However, with average lifetimes against deposition of roughly 4 to 10 days, most submicron aerosols that dominate number concentrations and production fluxes of primary marine aerosols were produced from biologically dis-

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tinct seawater far upwind of sampling sites. In addition, fresh aerosols undergo significant, rapid chemical modification involving the loss and accumulation of constituents via multiphase pathways. The text should be clarified.

Citations should also be added to identify the "field experiments" mentioned here.

Page 2284, lines 9-12. This statement suggests that all biogenic organic matter associated with marine aerosol in productive marine regions originates via direct emission from the surface ocean. Clearly this is not the case. For example, methansulfonic and oxalic acids are major organic constituents of aerosols in most marine regions but these compounds originate from secondary reactions involving biogenic precursors most of which are thought to be emitted as gases from the ocean surface. Anthropogenic and natural terrestrial hydrocarbons transported from continents are also important precursors for particulate organic carbon in many marine regions. The text should be clarified.

Page 2285, lines 1-4. What is a "typical marine aerosol size distribution signature?" This term should be defined or an appropriate citation added. Because lifetimes against deposition and vertical mixing potentials vary substantially as a function of particle size, size-resolved production fluxes are not directly comparable to size-resolved concentrations in ambient air. In addition, Sellegri et al. [2006] and Tyree et al. [2007] did not characterize the supermicron size fractions, which dominate the mass production flux and dry mass concentration of ambient marine aerosol and, as such, the cited distributions should not be described as "typical" of ambient.

Page 2286, line 2. The authors should clarify that sea salt dominates the non-water mass fraction of bulk marine aerosol. Typically, water dominates total mass in all size fractions of marine aerosol and organics often dominate the non-water mass of the smaller size fractions.

Page 2286, lines 3. O'Dowd et al. [2007] is not included in the references.

Page 2286, line 5. To minimize the potential for confusion, the authors should clarify

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that this statement refers to ambient submicron marine aerosol (to distinguish from freshly produced submicron marine aerosol).

Page 2287, section 2.1. It would be helpful to specify additional details such as (1) the orientation for the indicated dimensions of the tank (length, width, depth), (2) the range in flow rate of bubble air, and (3) the ranges in flow rate and velocity of the water jet.

In addition, bubble plumes in the surface ocean are typically 1 m or more in depth (see several papers by Thorpe and coworkers) and the organic enrichment of the primary marine aerosol increases with length of the bubble path [e.g., Hoffman and Duce, 1976, JGR]. The authors should point out the difference between their experimental design (i.e., bubble paths of 0.05 to 0.13 m and no organics) relative to conditions more typical of the surface ocean and discuss the potential implications for interpreting results.

Finally, the schematic in Figure 1 indicates that compressed air was processed through a HEPA filter upstream of the tank but HEPA filters remove only particles. The text (line 10) indicates that bubbles were generated and the chamber was swept with “zero air”, which is commonly defined as air from which both particles and reactive trace gases have been removed. The text and the figure should specify how reactive gases were removed. If they were not removed, the text should be corrected and implications for interpreting results discussed.

Page 2287, lines 17-20. Did the authors test the implicit but unstated assumption that a bubble plume produced by a single jet is representative of that produced by the same flow divided into eight jets? Some evidence suggests that this key assumption may not be valid [see for example Hoque and Aoli, 2008, Appl. Ocean Res.]. This issue and the associated implications for data interpretation should be addressed in the manuscript.

Page 2288, line 1. For experiments involving the sintered glass or diffuser, did 3 LPM correspond to the flow rate of bubble air, bubble air plus sweep air, or only sweep air? Presumably, for the water jet, this air flow was exclusively sweep air.

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Figure 1 indicates that sweep and bubble air were pushed into the tank and sample air simultaneously pulled from the tank. Was pressure maintained by precisely balancing the flow rates into and out of the tank or was the system operated under slight positive pressure and configured with a one-way exhaust valve in addition to the sampling lines. No such regulating device is shown in Figure 1. Since the analytical instruments require specific flow rates of sample air, presumably, some of the sample air must have been vented. If so, the vent should probably be added to the figure. If pressure was not regulated, was it monitored? If not monitored, how were potential artifacts resulting from variability in pressure evaluated?

Although specified in the text, it would be helpful to also define the major acronyms (BMS, CCD, MFC, etc.) in the legend.

Page 2288, lines 1-5. How and where was RH measured? Suggest adding to Figure 1.

Page 2288, line 5 to 20. Was the system blanked by analysis of sweep air in the absence of bubbles? If so, the results should probably be briefly mentioned. If not, how did the authors verify that the background was sufficiently clean?

Page 2289, line 1. The term “natural seawater” is misleading. The manuscript reports no experiments that were conducted using authentic natural seawater. It appears that all experiments were based on artificial seawater and that some experiment involved artificial seawater modified via addition of DOC produced by phytoplankton cultures grown under controlled conditions. The text should be corrected.

Page 2289, line 16. It would be helpful to briefly discuss the representativeness of organic-enriched artificial seawater produced in this way. Is it reasonable to expect that the physical and chemical characteristics of aerosols generated by bubbling air through these solutions were similar to those produced naturally at the ambient ocean surface?

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Page 2289, line 17. Was this procedure used to analyze the artificially generated organic material, the artificial seawater before enrichment with OC, and/or the artificial seawater after enrichment with OC?

Pages 2289 and 2290, Section 2.3. At what position (X, Y, Z) relative to the glass filter, diffuser, and jet was the BMS positioned for the various experiments? Why were these particular locations chosen? Later in the paper, the authors report that bubble size spectra generated with the water jet varied spatially within the tank but there is no mention of such variability for plumes generated with the other approaches or of variability as a function of depth. Were these evaluated? It would be helpful to briefly summarize in this section the specific locations within the tank at which bubble plumes were characterized for each production mechanism.

Given the very short distance between the bubble sources (5 to 13 cm below the surface for the glass filter and diffuser; 7 to 10 cm for the jet or jets) and the BMS (3 to 8.5 cm below the surface) coupled with the time delay between production and equilibration of the bubble plume [e.g., Lamarre and Melville, 1991, Nature], are these results considered representative of open ocean conditions? Some discussion regarding this important issue would seem warranted. Why was artificial rather than nature seawater used to characterize bubble plumes? Earlier in the manuscript (page 2284 lines 4-6 and page 2286 lines 10 to 15), the authors point out the potential the importance of organics in bubble plume dynamics and aerosol production. Did the authors characterize bubble plumes in seawater modified via addition of DOC? If not, why not?

Page 2289, line 25. Does the measured size range cover the full relevant size range of the bubble population within the tank? Most studies report upper limits for the bubble size distribution in the open ocean that are much greater than 1.1 mm diameter [e.g., see Lewis and Schwartz, 2004].

Page 2290, line 8. It would be helpful to specify the numbers of individual spectra that were averaged over 2 minutes.

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Page 2290, lines 10-13, and Figure 1. Most of the reported data for bubbles produced by the jet correspond to the BMS positioned at 110 mm from the center. Implications of the different positions of the BMS relative to the jet should be discussed here.

Page 2290, line 15. Suggest clarifying that only that "lower end of the aerosol size distribution . . ." was measured.

This section should also describe how the aerosols were transferred from the tank to the instruments. What were the measured or calculated transmission efficiencies as a function of size? It appears that the different experimental set ups were associated with different air fluxes. For example, those employing the glass filter and diffuser involved bubble rates of 5 to 10 LPM plus (presumably) 3 LPM of sweep air, plus 5% by volume of dry air whereas those employing the water jet corresponded to 3 LPM of sweep air plus 5% by volume of dry air. Did the apparent variability in the velocity of sample air through the lines influence transmission efficiency?

Page 2292, last few lines. Since organics influence surface tension and thus bubble plume dynamics, is there any evidence to support the implicit but unstated assumption that bubbles produced in OC-free, artificial seawater are representative of those in the surface ocean? If not, what are the implications for interpreting results?

Page 2293, lines 1-2, and Fig. 2. It would be helpful to depict and discuss the measured variability around these average distributions in the context of evaluating whether or not the differences in mean distributions were significant.

Also, here and elsewhere in the text, applying the term "aeration" to only the sintered glass and diffuser is ambiguous. The jet also introduced air into the test solutions.

Page 2293, line 19-20 and Fig. 4. The presentation seems out of order. Discussion of spatial variability in the bubble plume produced by the jet (as depicted in Figure 5) and the associated implications should preceded comparison of the plume produced by the jet with those produced by other methods.

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Page 2293, lines 24-28 and Fig. 4. Since bubble size distributions vary systematically as a function of depth [Lewis and Schwartz, 2004; and references therein] and age [Lamarre and Melville, 1991], it is not clear how a size distribution measured at 0.03 to 0.085 m depth can be directly compared with and interpreted in the context of older (on average) plumes measured at depths ranging from 0.5 to 7.3 m [Vagle and Farmer, 1992] and from 0.4 to 1.5 m [de Leeuw and Cohen, 2002]. This issue and associated implications for interpreting results should be addressed in the manuscript.

The terms for distance from the center of the tank and power-law exponents that are specified in the legend for Figure 4 should be defined in the caption. Here and elsewhere, to what does the plus sign preceding the distance from the center of the tank refer?

What is the rationale for selecting the data subsets depicted in Figure 4? More specifically:

- Why is the average distribution for the sintered glass at 10 LPM rather than at 8.4 for 5.2 LPM shown? Similar question for the diffuser. It appears that the distributions which diverged to the greatest degree from that for plunging water are depicted in the plot. What is the rationale for this?

- Why are size distributions for the jet that correspond to “quiescent” conditions (110 mm from the source) rather than “acoustic” conditions (at the source) compared with distributions for the sintered glass and diffuser that were apparently (based on Fig. 1) measured directly over the source? The size distribution for the plunging jet at the source (Fig 5) appears to be more similar to those for the sintered glass (at 5.2 LPM) and diffuser (at 8 LPM) than to the corresponding “quiescent” distribution for the plunging jet. Were distributions for the sintered glass and diffuser also measured away from the source under, presumably, more “quiescent” conditions? If so, those distributions would probably be more appropriate for comparison with those measured away from the jet. If not, the comparisons seem invalid since initial bubble plumes

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from any source evolve over time (and, in this case, over space). It appears that this analysis is based on the implicit but unstated and unjustified assumption that bubble plumes generated by jets evolve temporally whereas those generated by other methods do not.

- Why are bubble size distributions reported by Vagle and Farmer [1992] and de Leeuw and Cohen [2002] plotted whereas those reported by Dean and Stokes [2002] (cited later in the manuscript) or others [e.g., Bowyer, 2001, JGR; Terrill et al, 2001, JGR; Medwin and Breitz, 1989, JGR; Walsh and Mulhearn, 1987, JGR; among others] are not. An intercomparison of all such data would indicate a much greater range in variability than suggested in Figure 4 and the associated text.

- Both Vagle and Farmer [2002] and de Leeuw and Cohen [2002] report size distributions over ranges of depths and wind velocities. Which subsets of those data are depicted in Figure 4 and why were they selected?

- Why are the size distributions reported by Vagle and Farmer [2002] and de Leeuw and Cohen [2002] truncated at $\sim$600 μm? Should not be full size distribution be depicted in Figure 4?

- Finally, if organics are important in bubble plume dynamics as suggested earlier in the manuscript, it would seem more appropriate to compare bubble plumes measured in the open ocean with those generated in natural (as opposed to artificial) seawater or in artificial seawater amended with addition of DOC. I don't understand the rationale for ignoring the influence of organics in this component of the analysis.

The primary conclusion that the authors draw from Figure 4 (lines 26-28) is, in part, a function of the data subsets that were selected for plotting. A broader comparison would have yielded a considerably more ambiguous result. This important point should be stated explicitly and considered in the context of subsequent data interpretation.

Figure 5, caption and corresponding text on page 2294. The dashed lines and the

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power-law exponents should be defined.

More generally, did the bubble plumes generated with sintered glass and the diffuser vary at 55 and 110 mm from the center of the tank? Presumably, like those for the water jet, these plumes also evolved over time with distance from the source.

Page 2295, lines 5-10. Based on the above, this statement seems a bit strong. The reasonable correspondence may suggest that the plume evolved with distance from the source within the tank but it is certainly not a definitive indication that it reached a specific state comparable to an aged plume in the surface ocean.

What are the implications of this relationship for the aerosol generation experiments in which 8 jets, as opposed to a single jet, were used to produce bubbles? Presumably, multiple jets would correspond to a much larger area of acoustic conditions within the tank.

Figure 6. The legend should clarify that the size distributions correspond to particles less than 830 nm dry diameter. The larger sizes that dominate the mass flux and concentration were not characterized.

It appears that the upper end of the plotted size distributions for all but the atomizer are truncated at sizes smaller than 830 nm. Why is this?

Since each data point represents the average of 6 scans, it would be helpful to include the corresponding error bars to provide an indication of variability.

Page 2295, line 23, and Table 1. Should mode 4 be 341 nm as in the text or 340 nm as in the Table?

Page 2295, line 26. The terminology here is confusing. Mode 4 aerosol generated with sintered glass have a dpg of 253 not 340 nm.

Page 2296, lines 1-4. What is the basis for speculating on the importance of “splashing” as a source for mode 4 aerosol? In the absence of direct evidence, the statement as

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written seems overly strong. As mentioned in the preceding section of the manuscript, variability in the characteristics of bubble rafts (extent, thickness, etc.) also influences primary aerosol production. It is not clear how the relative influences of these and perhaps other factors could be differentiated based on data presented in the manuscript.

Page 2296, line 8-15. What does “stronger” splashing in this context mean? How is it quantified?

In the absence of evidence for the importance of “splashing” the discussion here seems rather speculative.

Finally, the authors have not reported spatial variability in the bubble spectra generated with the sintered glass or diffuser nor have they characterized bubble spectra generated by 8 jets (employed to produce aerosols) as opposed to a single jet. Consequently, there is little direct evidence that would justify the interpretation of aerosol production characteristics relative to the associated variability in bubble plume dynamics across the tank.

Page 2296, line 26 through page 2297, line 8. I don’t understand the logic here. As indicated above, submicron aerosols in ambient marine air are not representative of freshly produced aerosols at the ocean surface. The mass of dry submicron aerosol associated with on-shore flow at Mace Head is dominated throughout the year by non-sea-salt sulfate most of which originates from combustion sources over Europe [e.g., Savoie et al. [2002, JGR]. Even if freshly produced marine aerosols were chemically inert and there were no exogenous aerosol sources, size distributions for fresh versus ambient aerosols would still diverge in response to variability in deposition fluxes and associated atmospheric lifetimes as a function of particle size. Comparisons between ambient versus artificially generated aerosols such as those depicted in Figure 7 cannot be interpreted directly to evaluate the representativeness of the number size distributions or fluxes of aerosols produced at the ocean surface.

Page 2298, starting on line 11. It would be helpful to provide readers with some indi-

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cation of how the DOC concentrations used in this experiment compare to the range of those in ambient surface seawater.

Because the generator is a closed system, surfactant organics in the artificial seawater will be depleted over time via aerosol production and removal. Did the authors evaluate this potential complication in the experimental design via time-series measurements? How long could the system be operated before the depletion of surfactant organics began to influence aerosol characteristics? It would be helpful to mention in the methods section the duration of experiments and the frequency at which solutions were replaced.

Also, in a closed system, the delivery of organics to the surface by rising bubble can lead to the development of thick layers or organic foam that attenuate aerosol production. Did the authors observe evidence for such behavior? Were bubble rafts and/or foam more prevalent in the experiments involving OC-amended seawater?

Page 2301, lines 1-4. I don't understand this point. Bubble clouds produced by any mechanism are influenced by wake effects. Such influences are not limited to "aeration" methods.

Page 2301, lines 11-13. The description of QP is unclear. For sintered glass and diffusers, bubbles are generated by forcing compressed air through a porous medium not a single "pore".

The variables Vb and x should be defined.

Page 2302, lines 1-12 and Figure 10. It appears that this analysis is based on the implicit but unstated assumption that bubbles produced by a jet are not subject to wake effects whereas those produced by "aeration" are subject to wakes. What evidence supports this assumption? In a cloud, bubbles influence and are influenced by surrounding bubbles regardless of their mechanism of production.

Why is QP used in the equation but Q is used in the figure?

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Page 2302, lines 4-6. Which of the reported depths for jet plume penetration and the sintered glass location were used in this calculation? The Methods section indicates ranges of 7 to 10 cm and 5 to 13 cm, respectively.

In addition, presumably, the distance from the bottom of the jet plume to the surface corresponds to the approximate maximum rising distance for bubbles produced by the jet. Should not this analysis be based on the average or median rising distance for a "typical" bubble in the plume? How sensitive are the results to these distances?

Page 2302, lines 13-23. In the absence of measurements of the size-resolved organic content of the particles, this assessment seems rather speculative. At minimum, the statement regarding the size-resolved scavenging efficiency for organics should be supported with a citation.

Page 2302, line 24 through end of manuscript. The summary and conclusions require revision based on the above comments.

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