Due to some overlap between the reviewers' comments, both sets are combined below in one document.

Reply to Reviewers
Manuscript ID: amt-2016-115
Title: "Lake spray aerosol generation: A method for producing representative particles from freshwater wave breaking"

Comments from the reviewers are below with author responses in bulleted italics.

Anonymous Reviewer 1
General comments: This is an interesting and valuable study of aerosol production from bursting bubbles in freshwater, a potentially important subject that has been little studied. The authors made a thorough preliminary study of bubble and aerosol size distributions from lake water, synthetic freshwater, and synthetic seawater. I state "preliminary" not with any pejorative or diminutive implication, but merely that one could envision continued similar studies with different (and known) organic content, and especially different temperatures. Overall the manuscript is solid and I recommend publication. Before publication, however, there are a few minor topics listed below that should be addressed.

- *The authors appreciate this feedback and thank the reviewer for the helpful suggestions.*

Specific comments: Concentrations should be denoted in units grams/kg of water or grams/kg of solution (which, for seawater, is salinity), rather than in grams/L. To the number of significant digits given, the numerical values are probably the same, but stating values in grams/L (presumably L refers to water and not the solution) requires specification of temperature and (to a much lesser degree) pressure.

- *All concentration units previously noted as g L^{-1} of water are now expressed as g kg^{-1} of water.*

Abstract and throughout manuscript: Describing size distributions as having "a peak near 300 micrometer" is ambiguous in two ways: whether the size refers to radius or diameter is not specified (and for particles the RH to which this radius or diameter refers must be specified as well), and the peak is meaningful only if the representation of the size distribution is specified, i.e., whether the representation is in the form dN/dD or dN/dlogD.

- *Diameters were referring to diameter in $dN/dlogD_p$ space. The manuscript has been revised to clarify that sizes refer to diameter and are determined from log-normal distributions.*

The statement at the end of p. 4 that bubble coalescence is inhibited in seawater due to increased surface tension caused by higher ion concentrations is not correct. Yes, higher ion concentrations lead to inhibition of bubble concentrations, and higher ion concentrations lead to slightly higher surface tension, but attributing the cause of the increased bubble inhibition to higher surface tension is incorrect.

- *The authors have corrected this misstatement, and the manuscript now mentions the effect of ion concentration on bubble coalescence, but does not imply that surface tension is the cause.*

On line 10 on p. 5, the authors seem to stress that the bubble size distribution is the dominant factor controlling the resultant drop size distribution, but the concentration of species that could remain after water evaporation is perhaps a more important factor. Were freshwater to have the same bubble size distribution as seawater and drop production mechanisms were the same, the resultant drop size distribution would still differ considerably as the amount of material in the ejected drops that can remain to form the dry particles differs considerably between the two media.

- *The reviewer makes a great point and that sentence has been revised to reflect that bubble size is only one factor. Additional text referring to the impact of concentration on size is now on **Lines 17-23 of Page 5** and **Lines 1-2** of **Page 6.***

Toward the bottom of p. 7, the authors state that the depth (5 cm) is sufficient that it does "not affect the bubble plume or limit bubble lifetime," but this is not supported by any reference, and it would seem that breaking waves would entrain bubbles to more than 5 cm.

- *The authors have revised the previous statement about bubble plume and lifetime to reflect that, while not perfectly recreating ambient conditions, SSA generators (now cited here) have shown that small depths are effective at reproducing environment conditions and surfactant partitioning See **Lines 17-23** of **Page 8** and **Lines 1-8** of **Page 9.***

On p. 9 toward the bottom, the authors state that electrical mobility diameters and aerodynamic diameters "were converted to physical (geometric) diameters" but did not give any details as to how these conversions were done, the assumptions made, etc. This is important information that should be included.

- *Further information regarding conversion from electrical mobility and aerodynamic diameter to physical diameter is now included on **Lines 22-23** of **Page 10** and **Lines 1-12** of **Page 11**.*

On p. 17 the relation between solution concentration and bubble density was discussed, and different behaviors were seen for different ranges; thus why in Figure 7 was a linear fit assumed between these two quantities? Similarly, on p. 19 the nonlinear relationship between total aerosol concentration and solution concentration is discussed, but a linear fit is presented in Figure. 8.

- *The authors agree that linear fits should not have been used, and they have been removed.*

On p. 20 the authors conclude that low ionic concentration freshwater samples produce fewer particles than high ion concentration seawater samples, but the phrasing suggests that the ionic composition is somehow important rather than merely the greater solute concentration. To draw such a conclusion, it would seem that both ionic and nonionic solutions should be investigated. In contrast, on p. 21 they state that the low concentration of salts in freshwater is the reason.

- *The implication of composition playing a role has been removed and "ionic concentration" has been changed to "salt concentration" on **Line 21-22** of **Page 22** & **Line 22-23** of **Page 23**.*

In Figure 4, it is not clear if the dotted lines are lognormal distributions; this should be explicitly discussed. Additionally, it would aid the reader if a dotted line representing the sum of the two modes for the blue and red graphs were shown.

- *It is now explicitly stated that the distributions are lognormal and modes for each peak and the sum of the two modes are now included in **Figure 4** and the **Figure 4 caption**.*

In Figure 7, why are bubble size distributions displayed as dN/dD rather than dN/dlogD, similar to the representation used for aerosol size distributions? This would allow a more facile comparison between bubble production and drop production.

- *The authors agree and would prefer this, but chose to present as dN/dD to allow for direct comparison with prior SSA generation publications of bubble bursting (Fuentes et al., 2010; Hultin et al., 2010; Salter et al., 2014; Stokes et al., 2013), which are all dN/dD.*

In Figure 7, the labels differ from the caption (e.g., NaCl is shown in B).

- *The labels and captions have been corrected.*

Anonymous Reviewer 2

* General comments

This paper investigates Lake Spray Aerosol (LSA) through measurements of the size distributions and concentrations of bubbles and aerosol particles produced in an laboratory bubble generator. The measurements are interesting, comprehensive and generally well presented. The manuscript is nicely structured and written. The design of the LSA generator is nothing new. It is a standard plunging jet type bubble chamber. Instead the novelty of the work lies in the combined investigation of bubbles and aerosols generated in fresh- and salt-water solutions. I support publication in ACP after the following comments have been addressed.

The differences in the concentrations of aerosols produced from fresh and saltwater seems to be mostly explained by the corresponding differences in bubble concentrations. This is an interesting point, but as the authors have summarised nicely, the increase in the concentration of smaller bubbles in concentrated saltwater solutions due to the inhibition of bubble coalescence was first identified decades ago, and has since been observed in many different studies. I think there is an opportunity to use the combined dataset of aerosol and bubble size distributions to move beyond this point. For example, I suggest adding a plot (or plots) of aerosol mass concentration normalized by bubble concentration (or total bubble surface area or volume) against solution concentration. Fig. 1 indicates that the mass concentration of inorganic ions is ∼1000x less in fresh water than salt water. So it is surprising to observe that the total amount of aerosol produced from fresh water in the LSA generator is roughly comparable to the amount of aerosol produced from seawater (as far as I can tell from Fig. 4 taking the different bubble concentrations into account). How is it possible to account for this enormous discrepancy? The authors provide some possible answers in Section 3.2.3 in their discussion of droplet size distributions. Perhaps investigation of normalized aerosol mass concentrations will allow such discussion to be even more quantitative (i.e. the freshwater droplets would need to have ∼1000x the volume of the saltwater droplets to produce the same amount of aerosol mass).

- *Thank you for your suggestions. We have added a plot of the average total aerosol number concentration normalized by the average total bubble concentration as **Figure 4C**. As requested by the reviewer, we have also included here the plot of the average total aerosol mass concentration (from experimentally determined effective densities) normalized by the average total bubble concentration **(Figure A)** that illustrates as similar result to **Figure 4C**. We have chosen to include **Figure 4C** in the main text to maintain consistency in our discussion of aerosol size distributions and concentrations, and to provide for easier comparison with **Figure 4B**. **Figures 4C** and **A** demonstrate that the synthetic freshwater and seawater produce similar values for average total aerosol number and mass concentrations normalized to the average total bubble concentration. In comparison, the Lake Michigan freshwater sample produced a much larger average total aerosol number and mass concentration normalized to the average total bubble concentration. This difference can be attributed to the higher concentration of organic and biological material present in the L. Michigan freshwater. The effect of the organic and biological material present in the L. Michigan freshwater sample on aerosol size distribution and particle circularity was previously discussed in **Section 3.2.2** and further discussion in relation to Figure **4C** has been added to **Section 3.2.2** on **Lines 13-16** of **Page 15** and **Lines 17-22** of **Page 16**.*

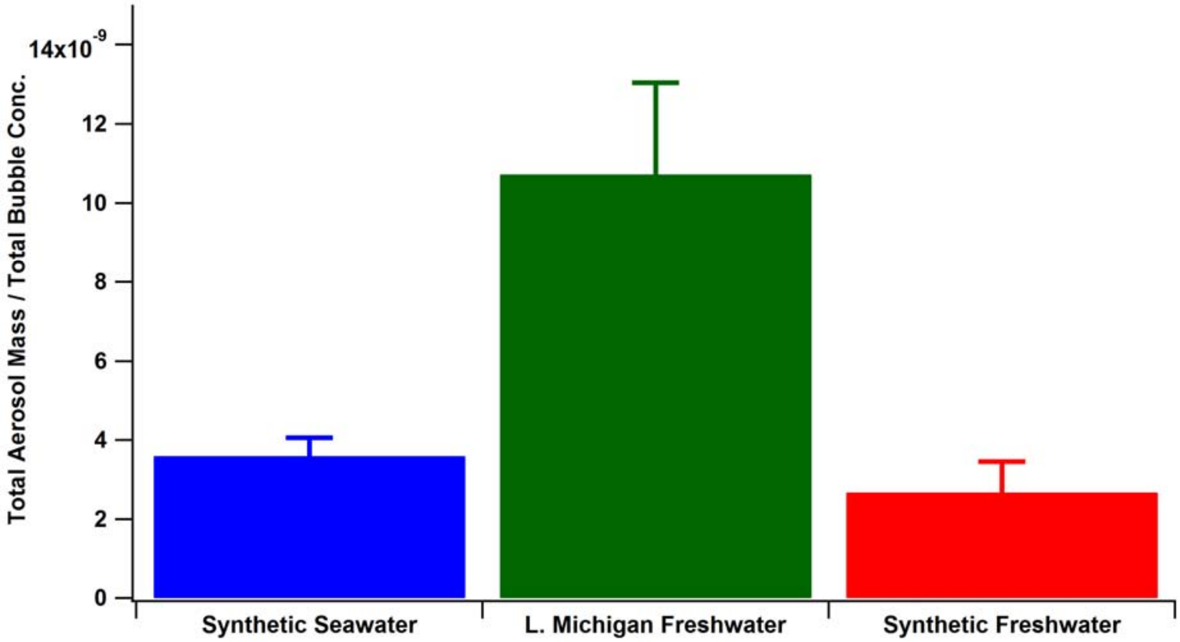

Figure A. *Average total aerosol mass concentration normalized by average total bubble concentration produced by the LSA generator from synthetic seawater, synthetic freshwater, and Lake Michigan freshwater.*

The citation and discussion of previous relevant studies is well done. I suggest to also add discussion of the early work of Monahan on freshwater whitecaps (Monahan, 1969; Monahan, 1971). Monahan observed that the wind speed threshold for freshwater whitecap formation is greater than the corresponding threshold for saltwater whitecap formation. He also observed that freshwater whitecaps decay faster than saltwater whitecaps. All things equal, these effects will result in less aerosol production from freshwater than saltwater. The broader point here, which is not made clear in the current introduction, is that the wind speed-whitecap relationship will likely be different over bodies of freshwater than seawater.

- *Text has been added to **Pages 3-4** that provides details (and the suggested references) about wind speed versus whitecap formation and lifetime over freshwater compared to seawater.*

** Specific comments

P2, L7: 'Larger bubble size distribution' is ambiguous (large sizes or concentrations?).

- *The text have been altered to clarify that this refers to the diameter of the mode.*

P3, L12: I would like to see some actual numbers from the Chung et al., (2011) study quoted here to indicate how much LSA might contribute to CCN concentrations. I think its important to state that even using an SSA source function, which should predict greater production of aerosol than a corresponding LSA function according to the results presented here, Chung et al., calculated that LSA only made minor contributions to total particle number concentrations. I think the manuscript overstates the climatic importance of LSA.

- *Numbers from Chung et al. (2011) have been added to **Page 3, Lines 4-8**. Since our manuscript submission, a publication on ice nuclei (IN) from freshwater (river) aerosol has been published and is now cited (Moffett, 2016). As a small number of IN can impact cloud properties and climate, the manuscript has been altered to reflect the potential climate impact of LSA as IN.*

P3, L23: The wind speed-whitecap (or -aerosol production) relationship will likely be different for bodies of freshwater than over the oceans. The fetch over lakes is smaller than over the oceans. The threshold wind speed for whitecap formation is greater over fresh water than salt water (Monahan, 1969).

- *The authors agree and more detail on freshwater whitecap formation is given on* ***Pages 3 & 4.***

P5, L14: The generator is fine for the purposes of this study, but I think it's too much to claim that the generator has been optimized for freshwater experiments. The only feature cited to support this claim is the small volume of the chamber, which is neither novel nor a unique aspect of freshwater experiments.

- *This is a good point. The phrase "with key features optimized for freshwater" has been removed.*

P7, L18: Can the authors cite any values for typical depth and size of breaking wave bubble plumes? It is important that the depth of the tank doesn't affect bubble plume depth and lifetime, but it is also important to know how the size of the plume in the LSA generator compares to real breaking wave bubble plumes.

- *Few studies on depth for freshwater plumes in situ are available and thus bubble lifetime in our generator may be affected by the dimensions. However, many studies of SSA with laboratory generators (of similar dimensions) produce comparable aerosol sizes and compositions to atmospheric and large scale chamber studies. Collins et al. (2014) describes in great detail that for a plunging waterfall the aerosol size and composition are comparable to wave breaking.* ***Lines 16-23*** *of* ***Page 8*** *and* ***Lines 1-6*** *of Page 9 now discuss these issues. As discussed in detail by Fuentes et al. (2010), the generation of plume depths >0.5 m by a plunging jet would be optimal for producing saltwater bubbles with a lifetime comparable to real plumes. However, this is not known for freshwater plumes, beyond tipping trough experiments that have their own limitations. This led us to use similar generator dimensions to those of validated SSA generators pending future ambient studies regarding freshwater plume depth and lifetime.*

P9, L15: Please provide a reference for these known quantification issues.

- *References describing issues with APS number concentration quantification in smaller bins have been added* (Ault et al., 2009; Khlystov et al., 2004; Qin et al., 2006).

P9, L18: What value(s) of effective density was used for this conversion?

- *Text has been added to* ***Lines 22-23*** *of* ***Page 10*** *and* ***Lines 1-12*** *of* ***Page 11*** *stating that experimentally determined effective density values ranging from 1.2 – 1.6 g* cm^{-3} *were used.*

P11, L6: The concentration of larger bubbles is an order of magnitude less than the Prather et al., (2013) measurements shown in Fig. 3, which are comparable to ocean wave measurements. Even if the decrease in the concentration of larger bubbles follows a similar power law as discussed on P12, L11, the absolute concentrations are still very small. This is a limitation of the LSA generator given the importance of larger bubbles as discussed in the next paragraph beginning on L8. This doesn't change the paper's conclusions, the measurements are all internally consistent, but I think the fact should be pointed out.

- *This limitation is now discussed directly on* ***Lines 9-16*** *of* ***Page 14****.*

P16, L12: Is it possible smaller bubbles were present even if they weren't measured?

- *Jet drops from smaller bubbles would have been large enough to observe, but were not observed in this study.*

P22, L9: Another interesting reference to add here is Woodcock's observations from 1948 (Woodcock, 1948).

- *A reference to Woodcock (1948) has been added to **Line 2** of **Page 25**.*

** References
Monahan, E. C.: Fresh Water Whitecaps, Journal of the Atmospheric Sciences, 26(5), 1026–1029, 1969.
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Woodcock, A. H.: Note Concerning Human Respiratory Irritation Associated with High Concentrations of Plankton and Mass Mortality of Marine Organisms, J. Mar. Res., 7, 56-62, 1948.