Response to Reviewer #2

The authors thank the reviewer for providing helpful and detailed questions.

Reviewer comments have been reproduced here in plain text, while author responses are provided in bold.

1. "A caveat should be stated upfront that while these laboratory studies are meant to mimic a breaking wave on the ocean and subsequent production of SSA, there may be other factors over the real ocean that are not being represented in the laboratory studies that control the flux and composition of SSA."

A statement to this effect has been added to the manuscript. The introduction of the wave breaking method in Section 1 now reads: "While laboratory waves may not reproduce all of the factors that lead to SSA production over the ocean, they do produce bubble size distributions that compare favorably with those measured in whitecaps (Deane and Stokes, 2002). Consequently, aerosol generation by the wave breaking method provides the closest proxy to natural SSA currently available in a controlled environment."

2. "Page 6460 line 10. Isn't this true at any wind speed?"

The phrase "at wind speeds less than 20 m s^{-1}" was meant to specify that bubble bursting is the only main driver of SSA formation at lower wind speeds. When winds increased to more than approximately 20 m s^{-1}, direct wind-driven spume droplet formation can be important (de Leeuw et al., 2011), and is not within the scope of this manuscript. This phrase has been omitted from the manuscript to avoid confusion.

3. "The ATOFMS measures particles down to 0.3 um Dva. Why do the chemical data in figure 2(a) go down to 0.1 um? Why don't the three plots cover the same size range? Most of the particle number is in sizes less than 1 um. Why are most of the ATOFMS counts in particles around 2 um?"

The cited size range of the ATOFMS with aerodynamic lens inlet on page 6466 (line 18) was mis-printed. The size range now reads d_{va} between 0.1-2.5 µm. The number size distribution sampled by the ATOFMS is dictated by the size-dependent transmission curve which is characteristic of the instrument's inlet (Gard et al., 1997; Su et al., 2004). For the ATOFMS with nozzle inlet, particles with d_{va} ~ 1.5 - 2 µm are sampled with greatest efficiency and as such account for a large fraction of the observed particles. Since transmission efficiency is size-dependent, the size-resolved chemical composition of particles is normalized to the total number of observed particles in each size bin. The number of chemically characterized particles with d_{va} < 0.5 µm is highly sensitive to the total number concentration of particles in that size range for each method. The sampling efficiency for particles with d_{va} < 0.5 µm is not as favorable as for particles with d_{va} > 1 µm. SSA generated by sintered glass filters is produced with number concentrations at the modal diameter of almost an order of magnitude greater than the plunging waterfall and breaking waves. The greater number concentration of SSA particles overall (and the smaller modal diameter) generated by the sintered glass filters allowed for chemical characterization of more particles, with a greater

probability of sampling particles with d_{va} < 0.5 µm. A similar description is now provided in the manuscript.

4. "It should be pointed out that the ATOFMS is looking at the tail of the number size distribution. That is clear from figure 6, but hidden in the log plot in figure 2. There is no information here about the chemical composition of most of the generated number population."

This observation was alluded to in the earlier version of the manuscript (p. 6471, lines 1-4) with reference to the trend in the CCSEM/EDX data at smallest particle diameters, but has been added to the manuscript in a more salient manner within the section concerning ATOFMS results and references the shape of the number size distribution of SSA particles.

5. "I assume the y axis in figure 2a is number fraction and not mass fraction. That should be made more clear."

This assumption is accurate. Since the ATOFMS measures particles individually, and not based on the total mass collected, the data are presented here in terms of number fraction. This also highlights the externally mixed nature of SSA. A note has been added to the caption of all figures to which this comment bears relevance.

6. "I realize there are uncertainties in converting between Dp, Dv, and Dva, but in comparing figure 2a and 2b, it would be nice to use the same particle diameter on the x axis."

In order to accommodate comparisons between the size resolved chemical composition and the number size distributions, a second horizontal axis has been provided for the ATOFMS data in which d_{va} has been converted to d_p. An accompanying discussion of the size metrics has been provided in the text, expressing caution in the use of d_p due to the various assumptions required.

7. "Is figure 4 the total size range of the ATOFMS?"

Yes. This has been clarified in the caption.

References

de Leeuw, G., Andreas, E. L., Anguelova, M. D., Fairall, C. W., Lewis, E. R., O'Dowd, C., Schulz, M., and Schwartz, S. E.: Production Flux of Sea Spray Aerosol, Rev. Geophys., 49, Rg2001,doi: 10.1029/2010rg000349, 2011.

Deane, G. B., and Stokes, M. D.: Scale dependence of bubble creation mechanisms in breaking waves, Nature, 418, 839-844,doi: 10.1038/nature00967, 2002.

Gard, E., Mayer, J. E., Morrical, B. D., Dienes, T., Fergenson, D. P., and Prather, K. A.: Real-time analysis of individual atmospheric aerosol particles: Design and performance of a portable ATOFMS, Anal. Chem., 69, 4083-4091,doi: 10.1021/ac970540n, 1997.

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