Response to Reviewer #1:

The authors thank the reviewer for constructive criticism and detailed comments. Changes made as a result of this review have strengthened the manuscript.

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

"They also demonstrated that although size-resolved aerosol composition differs among the techniques, the values for the bulk aerosol (i.e., averaged over all particles produced) was remarkably similar; a result I think they should highlight more prominently."

Indeed the striking difference between the averaged measurements and the size-resolved measurements was surprising, however it is consistent with the highly size dependent external mixing state of SSA particles, a concept that has been highlighted recently by several research groups (e.g., Prather et al., 2013; Deng et al., 2014; Park et al., 2014). The differences between averaged and size-differentiated measurements have now been highlighted in the abstract of the revised manuscript.

"Several different types of particles 'sizes' were used, but the authors were not careful about defining or distinguishing these."

The use of various size metrics has been clarified in the manuscript. In addition, a discussion of size distribution measurements and the size metric conversions from mobility (d_m) and aerodynamic diameter (d_a) to physical diameter (d_p) have been explained in Section 2, with relevant conversion equations and required assumptions. Caution has also been encouraged in the interpretation and cross-comparison of different measurements of particle size, based on uncertainties in the assumed density and morphology of the particles (e.g., p. 8, line 22-26, p. 9, lines 9-13)

p. 1, lines 22-24: these are two different thoughts (seawater properties vs. production mechanisms); conclusion doesn't follow logically

These two concepts have been distinguished, yet linked through revision. The passage now reads: "Bubble bursting is sensitive to the physicochemical properties of seawater. For a sample of seawater with constant composition, any important differences in the SSA production mechanism are projected into the composition of the aerosol particles produced.

p. 2, line 2: suggest change "than sintered" to "than those produced by sintered"
p. 2, line 2: suggest change "generated by disintegrating foam produced by" to "from"
p. 2, line 3: "organic enriched" should be "organic-enriched" as in line 9
p. 2, line 11: suggest "compared to" to "compared to those produced by a"
Each of these changes has been made.

p. 2, line 4: "size" is ambiguous and has not been defined; does it mean radius or diameter, and at what RH?

This reference to size has been changed to 'dry diameter'. The sampling RH is now given in the first sentence of Section 2.3.

p. 2, line 15: do you mean "aerosol" or "aerosol particles"?

Essentially, either of these terms is valid in this case – the results of this study show that the internal mixing state of individual aerosol particles is different to some degree for each of the methods, while the external mixing state of the aerosol population is also influenced by the SSA production method. The statement now reads: "Throughout this set of experiments, comparative differences in the SSA number size distribution were coincident with differences in aerosol particle composition, indicating that the production mechanism of SSA exerts important controls on both the physical and chemical properties of the resulting aerosol with respect to both the internal and external mixing state of particles." In addition, a sentence introducing the concept of external and internal mixing state has been added to Section 3 preceding the description of the chemical analysis by ATOFMS.

p. 2, lines 15-17: it is not physicochemical differences between mechanisms, but between particles produced by different mechanisms; suggest reword accordingly.

The purpose of this statement was to highlight the intimate connection between the physicochemical properties of the seawater, the bubble bursting process, and the resulting sea spray aerosol. The sentence now reads: "This study provides insight into the inextricable physicochemical differences between each of the bubble-mediated SSA generation mechanisms tested and the aerosol particles that they produce, and also serves as a guideline for future laboratory studies."

p. 2, line 28: "at wind speeds less than 20 ms-1" contributes nothing; suggest omit
p. 3, line 1: suggest change "salt" to "inorganic salts"
p. 3, line 3: suggest change "aerosol is enhanced when ejected" to "aerosol particles is enhanced when the particles are ejected"; as it reads now, the organic fraction is what is ejected
p. 3, line 18: suggest change "only" to "primarily"
Each of these changes have been made.

p. 3, line 19: not clear what is meant by "traditional atomization methods"

The text has been changed to "pneumatic atomization" to draw a distinction from bubble bursting aerosol generation methods.

p. 3, line 22: Monahan and Zietlow (1969) and Cipriano and Blanchard (1981) also used an impinging water jet, and and Cipriano et al (1983) also used a frit; this may not be relevant as you mention laboratory studies of aerosol composition, but you do list Cipriano et al. (1983)

The cited works here are now more precisely restricted to studies specifically investigating nascent SSA composition.

line 29: as sentence reads, the bubble size distribution is the best proxy to natural SSA; suggest reword

The sentence 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."

p. 4, line 15: suggest that you define foam; it has been used with various meanings in the literature

The term 'foam' has now been defined in the second sentence of Section 3.2, which discusses influence of foam on SSA composition and number size distribution. The sentence (in Section 3.2) reads: "Natural SSA is mainly produced by whitecaps in the ocean, which are episodic in nature (de Leeuw et al., 2011). The visible white area on the sea surface during and subsequent to a wave breaking episode is due to the presence of foam, a collection of bubbles floating at the air-sea interface, each separated from the next by a thin liquid film (Bikerman, 1973)."

p. 5, line 23: suggest give water volume flow rate as you did on p. 6, line 20

The water volume flow rate for the plunging waterfall inside the wave channel is not available.

p. 5, line 30 and p. 5, lines 1-2: this list is nearly identical to that on p. 3, lines 22-24; suggest omit entire sentence, as you have stated this information on the previous page

This sentence has been omitted.

p. 6, line 6: state Keene's value (rather than merely "which is smaller than that used by Keene")

The value for bubble rise distance reported by Keene et al. (2007) has been included in the text.

p. 6, line 21: by the statement "from the ocean surface 275 m offshore" is it to be inferred that this is a different from that stated on p. 4, line 26 of "approximately 4 m below the low tide line"?

Yes. The source of seawater for the wave channel is sampled at depth through a seawater delivery system that is part of the infrastructure at Scripps Institution of Oceanography as described in detail by Prather et al. (2013), while the Marine Aerosol Reference Tank was supplied with seawater from the ocean surface, sampled manually. A comment has been added to the sentence in question to draw this distinction.

p. 6, line 31: over what time period?

The phytoplankton grow over a period of 1-2 weeks. This has been added to the manuscript.

p. 8, lines 1-2: as d_va depends on density (and shape), details of how this translation was done are necessary

The value of d_{va} is based on the measured velocity of particles traveling between the continuous wave lasers, as described in the manuscript. A calibration curve is constructed relating the velocity to the known d_p of polystyrene latex (PSL) spheres, which have a well-defined density and shape. This calibration curve is then applied to the velocity of each SSA particle. As a convenience to the reader when comparing the size-resolved composition to the number size distributions, a second horizontal axis for the ATOFMS data has been provided, showing the conversion of d_{va} to d_p (DeCarlo et al., 2004) for spherical particles with an assumed density of 1.8 g cm^{-3} (Zelenyuk et al., 2007).

p. 8, line 12: suggest change second instance of "based on" to "according to"

This change has been made within the manuscript.

p. 9, line 5: suggest title be descriptive and not conclusive (e.g., "Influence of sea spray generation method on SSA composition")

The header to Section 3.1 has been changed to: "SSA generation method inter-comparison and particle composition".

p. 9, line 11: suggest change "production" to "produced" as it is not the characteristics of production that you discuss

This sentence is not referring to the present study, rather it is referenced to prior studies that highlight the influence of seawater physicochemical properties (e.g., surface tension, temperature) on the production of SSA.

p. 9, lines 14-16: sentence is out of place here; suggest omit

This sentence was revised to draw a connection between the discussion of prior studies and the results of the current study. It now reads: "This study extends prior investigations by directly probing the influence of bubble bursting mechanisms on the detailed composition of SSA particles using direct chemical measurements at the single particle level."

p. 9, line 23: here you group into four types, whereas on p. 8, line 10 you state five types; suggest clarify

The fifth type is a group of particles labeled "other". This has now been clarified in the text: "A fifth type labled 'Other' contains Art-2a clusters with minor contributions and/or are attributed to contamination from lab air."

p. 9, lines 26-27: supermicron and submicron are ambiguous terms – do these refer to aerodynamic diameter? If so, presumably this is at 0% RH? It would be better to state d_va values rather than use terms such as these

All aerosol sampling was conducted at RH < 15%, which has been added to Section 2.3. All mentions of 'supermicron' and 'submicron' have been changed to $d_{va} > 1$ and $d_{va} < 1$, respectively, noting the now careful use of d_{va}, d_{ae} (area equivalent diameter), and d_p throughout the revised manuscript.

p. 9, line 29: why in Figure 2 does the value of d_va for wave breaking extend only down to 0.4 um? The number concentrations are still high at values below this

The sampling characteristics of the ATOFMS are dictated by the size-dependent efficiency of the inlet. The efficiency for $d_{va} > 1$ µm is sufficient that a fair number of particles could be sampled in all cases. For $d_{va} < 0.5$ µm, the number of particles that were chemically analyzed in each size bin scales with the number concentration produced by each method within each size bin interval. The sintered glass filters produced about a factor of 10 more particles than the other two methods, hence more particles with $d_{va} < 0.5$ µm were observed.

p. 10, line 2: "were" should be "was" as it corresponds to the subject "number"
p, 10, line 9: suggest "mechanism" rather than "mechanisms"
p. 10, line 10: "single particle" should be "single-particle"

These changes have been made.

p. 10, line 12: presumably diameters are d_proj; suggest that you state explicitly

The equivalent projected area diameter label d_{proj} has been replaced with equivalent area diameter (d_{ae}), with no change in definition. The diameter metric for electron microscopy data has now been explicitly defined in all relevant instances.

p. 10, line 24: suggest change "in submicrometer particles" to "in particles with d_proj < 0.8 um"

This change has been made.

p. 10, line 31: suggest omit sentence after "measurements" as this is contributes little and weakens your argument

The second part of this statement stresses the physical reality of why such specific measurements are useful for aerosol particles, rather than simply asserting that the measurement technique is important. The text has been revised slightly to be more concise: "This disparity between the two means of analysis highlights the importance of making size-resolved single particle measurements due to the inherent chemical heterogeneity in an externally mixed aerosol."

p. 11, line 4: suggest change "concept" to "tendency" or something less vague

This change has been made.

p. 11, line 8: "what "have been shown"? – subject of sentence is "concept"

This statement has been split into multiple sentences for clarity. "The tendency for inorganic cations to coordinate, or strongly associate, with organic and biological material in the ocean and in SSA particles has long been recognized (Duce and Hoffman, 1976). Divalent cations, such as Mg^{2+} or Ca^{2+}, have the ability to stabilize organic supramolecular structures (Verdugo, 2012) and coordinate surface active ligands at interfaces (Casillas-Ituarte et al., 2010). Magnesium has been shown to be a good tracer for SSA produced from bacteria-rich seawater (Guasco et al., 2013) and has been observed in aerosol over the ocean in association with biological activity (Gaston et al., 2011)."

p. 11, lines 13-14: this is a repeat of p. 10, lines 24-25; suggest omit

This statement has been revised to be less repetitious, yet still provides a link the the final important statement of the paragraph: "The depletion of Mg, K, and Ca in particles with d_{ae} < 0.8 µm observed by CCSEM/EDX suggests that smaller particles have chemical trends opposite those with d_{ae} > 1 µm."

p. 11, line 23: "do" should be "does" as the subject is "fraction"
p. 12, line 6: suggest change "diameter" to "values of d_proj"

These changes have been made.

p.12, lines 13-16: a table of the fraction that are spherical and the fraction that are cubic for each of the three bubble production mechanisms would be helpful

The manner in which we report the circularity findings is sufficient. Less than 1% of particles were not found to satisfy the requirements for either 'spherical' or 'cubic', so roughly the remainder of the particles not cited as spherical were cubic.

p. 12, line 26: suggest remove periodic; perhaps replace with episodic

p. 12, line 28: "are" should be "is" as subject is "nature"

These changes have been made.

p. 13, line 15: here Dp used which is not defined – at what RH is this measured?

The sizing metrics used for number size distributions are now explained in detail in Section 2.3. The metric used is the physical diameter (d_p), which allows APS and SMPS data to be merged into one size distribution.

p. 13, lines 22-23: this is a repeat of line 15 above and thus should ber removed

This has been removed.

p. 13, line 24: suggest not using "ultrafine" and "coarse" as these terms mean different things to different people, but instead stick with more quantitative descriptions (i.e., Dp)
p. 13, line 28: similarly with "accumulation mode" as this refers to atmospheric aerosols that typically attain this size through cloud processing

All size classifications are now identified using specific numerical ranges, including the relevant size metric.

p. 14, line 28: Figure 8 doesn't really contribute anything meaningful; suggest omit

Figure 8 provides a pictorial reference for readers who are less oriented with how the bubble bursting process progresses, including the various factors that lead to chemical differences in the SSA produced by different methods (implying different physicochemical scenarios). This figure has been retained in the revised manuscript.

p. 15, lines 22-23: it is highly likely that during wave breaking the SML would be disrupted, so this argument does not seem valid

The SML has been shown by Wurl et al. (2011) to exist in the open ocean with winds up to 10 m s^{-1}. Wave breaking can initiate around 5 m s^{-1} (de Leeuw et al., 2011), which suggests that the SML does exist when wave breaking is active. Taken together, Section 4 of this manuscript discusses the several processes which control the partitioning of organics to SSA (via the SML). For instance, the section on mixing discusses the concept that surface active organics at the surface of the water could be mixed back down into the water column by breaking waves. This mixing likely limits, but does not necessarily preclude, the formation of an SML.

p. 16, lines 6-9: this belongs in the experimental section

The penetration depth of bubbles has been transferred to the experimental section. This passage now reads: "While this study did not systematically test the bubble transit distance as a contributing factor in SSA organic matter content, we note that the transit distance of bubbles from breaking waves in this set of experiments (which includes a downward and upward path) was similar to that of the sintered glass filter bubble plume (upward path only), since the penetration depth of the wave was about half the depth of the sintered glass filter setup."

p. 16, line 12: should be "counteract" with no hyphen

p. 16, lines 20-23: same as p. 15, lines 17-19; suggest remove one instance

This change has been made.

p. 17, lines 25-28: it is not clear how these two options differ; also, are these the only possibilities?

These two hypotheses are not mutually exclusive, nor are they the only possibilities. This has been clarified in the revised manuscript.

p. 18, line 4: "lack" implies complete absence, which is difficult to determine from the figure – where there absolutely NO particles, or merely low concentrations (in which case "lack" is not appropriate)?
p.; 18, lines 4-5: film drops are also produced in this size range – where are they?

This sentence has been revised: "The low concentrations of SSA particles with dp > 0.3 um is perhaps indicative of reduced aerosol production by the jet droplet mechanism, based on particle size (Lewis and Schwartz, 2004; de Leeuw et al., 2011).

p. 18, lines 11-13: the conclusion that all particles with dp > 0.3 um are jet drops has not been established, although you are taking this as true

Recognizing this fact, this sentence has been revised: "In either case, the reduced influence of particles with d_p > 0.3 µm to SSA when a persistent layer of foam was observed clearly links a change in SSA production with surface foam accumulation."

p. 18, lines 23-24: more the mechanism than the physicochemical environment

The physicochemical environment in which a bubble bursts is controlled by the bubble production mechanism though each of the processes outlined in Section 4. The stark difference between the composition of SSA from sintered glass filters and those produced by wave breaking help support this concept. If the physical and/or chemical properties of the seawater in which the bubbles were bursting was not different in the local environment around the bubble, a chemical difference in the SSA could not have been observed. The connection between the bubble production mechanism and the physicochemical environment has been made in Section 5.

p. 18, lines 27-29: as noted above, this conclusion is not justified, as the drop type was never established

This sentence has been revised to accentuate the greater influence of thin fluid film rupture to SSA production in the presence of thick foam. The text now reads: "Based on the results presented in Section 3.2, it is also shown that SSA production studies with accumulated foam layers can be a useful tool in enhancing the formation of SSA via thin fluid film rupture."

p. 19, lines 1-3: no figure is associated with these values – were they determined from the size distributions shown in Fig. 2? This should have been discussed earlier and not for the first time in the conclusion section

These values for modal diameters were moved to Section 3, with an accompanying description prior to the presentation of chemical analysis results.

p. 19, lines 6-7: suggest reword as "contribution of OC from . . . particles"

This change has been made.

p. 19, lines 8-10: also very important would be the bubble size distribution difference resulting from the production mechanisms (as determined by the sizes of frits used)

The importance of the bubble size distribution has been established by prior studies. This sentence was meant to highlight new influences elucidated by this study. The sentence in question now reads: "While it has been established that the similarity of the bubble size distribution in laboratory SSA generators to that in the open ocean is an important factor (Fuentes et al., 2010; Prather et al., 2013), these authors suspect that turbulent mixing of the organic-enriched surface microlayer back into the water column by the breaking waves and plunging waterfall, and perceived lack thereof by the sintered glass filters (as implemented in this study), may play an added role in the results presented in the inter-comparison portion of this study (Section 3.1)."

p. 19, Appendix A: As this is important material and not too long, I would suggest putting most of it in the main part of the manuscript. Lines 1-7 on p. 20 can be omitted, as the reader is referred to Prather et al. (2013), which describes the technique, but lines 8- 26 on p. 20 comprise one paragraph that would fit in nicely above.

The material in the appendix has been incorporated into the main text as suggested.

line 20, Appendix B: Similarly, as this is only one paragraph, it can easily be incorporated into the main text.

Appendix B has been omitted, and the table has been re-named Table 1.

p. 28, Fig. 2: caption to lower axis on right-hand figure: Dp is mobility diameter, not physical diameter

The physical diameter is used in this figure as a way to combine the APS and SMPS data. As noted above, a description has been added to the main text.

p. 29, Fig. 3: caption to lower axis should specify which type of diameter (probably

Dproj)

The axis has been re-labelled as "Area Equivalent Diameter (d_{ae}; µm)"

p. 29, Fig. 3 insets: these are small and values are close to unity; suggest making y-axis range from 0 to 1.5 or 0 to 2 rather than 0 to 10 to aid the reader

The vertical axes on the insets to this figure have not been changed to draw the reader's eye to the stark differences between the averaged and size-resolved data.

p. 32, Fig. 6: suggest have dotted line at number concentration = 0 so that reader can determine how close to zero values are at larger diameters

A dashed zero line has been added.

p. 32, Fig. 6: specify which diameter (presumably Dp, mobility diameter)

Physical diameter has been specified.

p. 34, Fig. 8: as noted above, suggest omit as this contribute nothing meaningful

A response to this comment has been given above.

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