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Atmospheric Measurement Techniques Discussions

Interactive comment on "Direct aerosol chemical composition measurements to evaluate the physicochemical differences between controlled sea spray aerosol generation schemes" *by* D. B. Collins et al.

Anonymous Referee #1

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The manuscript describes clever and careful experiments performed in a laboratory wave tank comparing chemical composition of individual sea spray particles produced by three different methods using actual seawater: a breaking wave, a waterfall, and by bubbling air through frits. The experiments provide valuable insight into the extent to which the different methods agree, and demonstrate conclusively, using a variety of chemical composition characterizations, that frit production is not a good proxy for natural wave breaking. They also demonstrated that although size-resolved aerosol composition differs among the techniques, the values for the bulk aerosol (i.e., aver-

aged over all particles produced) was remarkably similar; a result I think they should highlight more prominently. They also performed experiments with a pulsed vs. a continuous waterfall to investigate the chemical composition of aerosol particles produced from the two techniques and the dependence of this composition on the occurrence of a continuous foam layer.

The paper should be published, and it will be a valuable contribution to the literature. There are a few issues that should be addressed, but these are relatively minor. Several types of particle "sizes" were used, but the authors were not careful about defining or distinguishing these. In some instances the manuscript is repetitious, and in quite a few the subject-verb agreement is incorrect (although these don't detract from the scientific contributions, they make the manuscript difficult to read). Some of the conclusions do not seem justified, and that section could be tightened up a bit, as it doesn't seem to have a clear focus. I have included a (long) list of suggested changes that I think would make the manuscript stronger and easier to read; mostly these involve tightening up the writing and should take little time to do.

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

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

p. 2, line 11: suggest "compared to" to "compared to those produced by a"

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

p. 2, lines 15-17: it is not physicochemical differences between mechanisms, but be-

tween particles produced by different mechanisms; suggest reword accordingly

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"

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

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

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

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

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

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

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"?

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

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

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

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

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

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

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

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

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

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"

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

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

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

and weakens your argument

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

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

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

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"

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

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

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

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

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

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

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

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

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

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

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?

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

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

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

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

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

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)

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.

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

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

physical diameter

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

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

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

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

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

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Interactive comment on Atmos. Meas. Tech. Discuss., 7, 6457, 2014.