

We would like to thank both reviewers for their constructive suggestions, which have significantly contributed to improve the quality and clarity of our manuscript. We carefully considered each comment and have made the necessary modifications accordingly. Specific responses to the reviewers' comments, along with the corresponding changes in the manuscript, are provided below in red text.

Reviewer 1

General comments:

This paper is written on the development, characterization, and application of a marine aerosol generator. The authors provide a very thorough review of relevant literature both in the introduction and throughout the manuscript. Multiple tests were conducted to validate the results of their system, while also comparing to results of similar systems. The system is then applied to several samples to compare their size distributions, chemistry, and INP concentrations. The results are thorough and make for a linear story as well.

A/ We sincerely appreciate the reviewer's thorough and detailed assessment, as well as her/his positive feedback on the manuscript.

Specific comments:

- Lines 164-166: Cite or mention Stokes et al. as reference for system

A/ Thank you for the suggestion. The following sentence was added to the revised manuscript:

Lines 167: "similar to the Stokes et al. (2013) tank"

- Line 178: Provide brief explanation on why the chosen cascade was the most suitable.

A/ We thank the reviewer for the suggestion. The following text was added to the revised manuscript:

Lines 180-181: "Other cascades were tested; however, the commercial cascade was selected as it generated the highest concentration of aerosol particles"

- Are OPC counts biased towards the lowest bins?

A/ The used OPC is able to measure aerosol particles between 0.3 and 10 μm . As shown below in Figure A1 from Prather et al. (2013), the particle size concentration around 0.3 μm is always higher than larger particles. Based on the results from Prather et al. (2013) and other studies focusing on laboratory and ambient marine aerosols, we are convinced that the behavior of our OPC is correct, i.e., there is not a bias towards lower sizes.

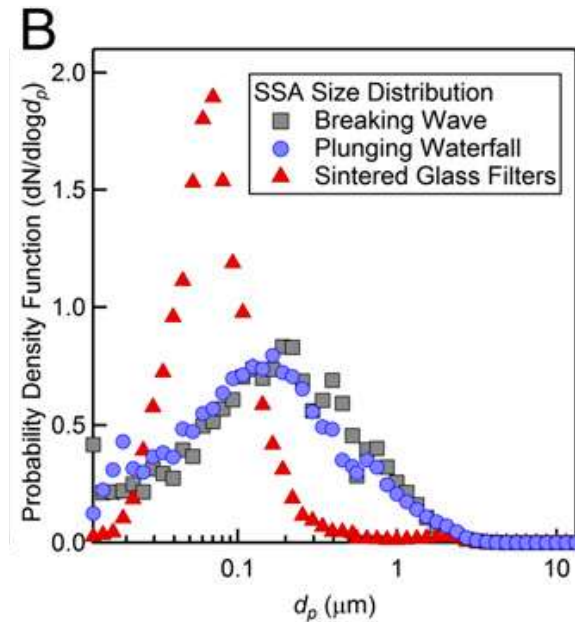


Figure A1. Probability density function of the resulting SSA number distributions ($dN/d\log d_p$, with the d_p at $15 \pm 10\%$ RH) produced by three different methods (for breaking waves (gray squares), plunging waterfall (blue circles), and sintered glass filters (red line)). Modified from Prather et al. (2013).

- 3.1: Which cascade was used for the background particle test?

A/ The results shown in Figure 3 were obtained using the commercial cascade. The following sentence was added to the revised manuscript for clarity:

Line 311: “water with the commercial cascade off (black line) and with the commercial cascade on (blue line)”

- 3.1: At what (time) point does the SSA reach a steady state? This information would be useful.

A/ The steady state was typically observed after 20 minutes. To acknowledge this, the following text was added to the revised manuscript:

Lines 209-211: “The samples were collected after 20 minutes of aerosol generation given that this time was set as the point where SSA reached a steady state.”

- 3.2: Which cascade is the best/most realistic? Should be explicitly stated here.

A/ Thank you for the observation. As seen in Section 3.2, cascades C and D showed the highest concentration. However, cascade D was chosen for the subsequent experiments because its standard deviation was slightly lower than that of cascade C. Additionally, Cascade D was easier to clean which was helpful to avoid interferences in later experiments. Cascade C has inner holes, which might have made the cleaning

procedure more difficult and uncertain when working with samples collected from the ocean.

The following sentence was added to the revised manuscript:

Lines 345-347: “Cascade D (the commercial one) was selected for the subsequent experiments because it produced the highest particle concentration, its standard deviation was slightly lower than cascade C and it was the easiest to clean”.

- It should be mentioned how the longer delay also allows for resettling/reformation of the SML, which is important for the chemistry of the particles.

A/ We thank the reviewer for the suggestion. The following text was added to the revised manuscript to provide a better discussion.

Lines 383-385: “However, it is important to note that the longer delay also allows for reformation of the SML, which is important in the composition of the marine aerosol”

- Line 179 states that with 40 L of water, the waterfall is roughly 10 cm, however Lines 401-402 state that a 40 L fill results in a 22.5 cm waterfall height. This is a 2x difference. Please address this discrepancy.

A/ Thank you for detecting this unintentional mistake. The correct height is 22.5 cm, therefore, this value corrected in **Line 182** in the revised manuscript.

- Line 464-465: Not necessary to state here that coarse mode particles can be produced. This point is made and should be stated in section 3.5

A/ We agree with the reviewer. Therefore, the sentence was moved to Section 3.5 in the revised manuscript.

Lines 439-440: “Additionally, it is important to highlight that the UNAM-MARAT can produce coarse-mode particles ($> 1 \mu\text{m}$).”

The information in the Section 4.1 was slightly modified.

Line 486: “Generally, the coarse particles correspond...”

- The authors reference how similar works induced phytoplankton blooms and that is what accounts for varying INP concentrations in other works. A comparison of INP concentrations from their work to INP concentrations from the start of the blooms (prior to culture additions) for other works would be a more appropriate comparison and useful addition.

A/ Thank you for this valuable suggestion. We acknowledge the importance of comparing INP concentrations from our work to those measured at the start of phytoplankton blooms in similar studies. A brief discussion was added as follows.

Lines 564-570: “When comparing our results with the abovementioned studies (i.e., McCluskey et al. 2017; DeMott et al. 2016) before the addition of the culture medium (Day 0), we find that our INP concentrations are rather comparable with the values reported by both studies. However, the clear difference between the former studies and our results is that more efficient INPs were observed during the bloom conditions, as they nucleate ice at warmer temperatures (i.e., $> -15^{\circ}\text{C}$), a situation not observed in our study.

- Centrifugal pumps can be harsh on biology and this could be affecting overall INP concentrations. Have authors considered this and measured if the number of INP at specific temperature changes over time? Another option is to perform experiments with other pumps to test affect on INP concentration.

A/ Thank you for your comment and suggestion. We agree with the reviewer that this could impact the results; however, there it is not a clear consensus in the community. For example, Lee et al. (2015) discussed how pump-induced cell lysis could inhibit the phytoplankton growth. However, in their mesocosm experiments phytoplankton growth was stimulated using a growth medium until the Chl-a threshold was reached. At that point, the plunging waterfall was activated to generate SSA. Even after the operation of the mechanical pump for SSA generation, the authors observed that some phytoplankton species remained alive and continued blooming, as indicated by the increase in chlorophyll concentrations for several days after aerosol generation. On the other hand, intact cells are not necessarily required to act as INPs, as cell fragments or their exudates can also exhibit ice nucleation abilities as shown by Wilson et al. (2015).

In conclusion, although the usage of the centrifugal pump may have impacted the INP concentrations, unfortunately we did no evaluate its influence. However, we will try to check this out with the UNAM-MARAT in a follow up study where marine biology will be carefully monitored.

To acknowledge this, the following text was added to the revised manuscript:

Lines 644-646: “However, it is important to note that the usage of a centrifugal pump could impact the marine microorganisms present in the natural seawater samples, potentially affecting their ice nucleating abilities”.

Technical comments:

- Line 74: Delete “in.”

A/ Deleted

- Line 120: Suggest using “species” instead of “particles” as most particles can contain more than just a diatom or dinoflagellate.

A/ Corrected

- Line 190: Water flow or air flow?

A/ In this case, the flowmeter measured the water flow. No change was applied.

- Figure 1: This is meticulous, but only 8 screws are accounted for in diagram and text states that 10 are used to hold lid.

A/ Good eyes, thank you. Figure 1 was corrected in the revised manuscript.

- Line 275: Section 2.4 label

A/ Corrected

- Line 334: *produced

A/ Thank you. This was fixed.

- Line 481: Should be decreases, not increases

A/ Thank you. The word was changed.

- Line 613: *tank

A/ Thank you. This was fixed.

References

DeMott, P. J., Hill, T. C. J., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., Hwang, C. Y., Rhee, T. S., Snider, J. R., McMeeking, G. R., Dhaniyala, S., Lewis, E. R., Wentzell, J. J. B., Abbatt, J., Lee, C., Sultana, C. M., Ault, A. P., Axson, J. L., Diaz Martinez, M., Venero, I., Santos-Figueroa, G., Stokes, M. D., Deane, G. B., Mayol-Bracero, O. L., Grassian, V. H., Bertram, T. H., Bertram, A. K., Moffett, B. F., and Franc, G. D.: Sea spray aerosol as a unique source of ice nucleating particles, *Proc. Natl. Acad. Sci.*, 113, 5797–5803, <https://doi.org/10.1073/pnas.1514034112>, 2016.

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