

This manuscript describes a thorough analysis of the life cycle of DOC concerning concentrations in seawater, the SML, and integration into SSA during a phytoplankton bloom. Using various methods, the concentration and enrichment of DOC across these layers of the air-sea interface are quantified and related to the biological activity and interactions occurring. Namely, the non-linear relationships between aerosolized DOC and chlorophyll concentrations point to complex interactions leading to issues with current limitations on understanding the DOC aerosolization cycle.

### **Major Comments:**

- The figures with timeseries are misleading due to inconsistent time intervals. It is recommended to update the figures with consistent and evenly distributed intervals to better present changes and make it easier to compare between plots when measurements were taken on different days and at different intervals.
- I have a few concerns about your experimental setup and description. Firstly, were the carboys aerated or sealed? From Fig. S1, it appears they were sealed and not aerated, which would limit the type of community that thrives during the experiment. Secondly, I appreciate the updated main text concerning the air temperature fluctuations of the carboys. However, the text states the average temperature was 21C, while the SI text mentions the experiments being conducted at 25C. Can you explain this discrepancy and if the 25C is the water, aerosol stream, or room temperature? Thirdly, you mention operating the simulation tank on continuous waterfall mode with a centrifugal pump for 8-9 hours. How did you address the time-dependent destruction of the microbial community due to the centrifugal pump? What period of those 8-9 hours did you use to estimate the size distribution? Fourthly, what do you mean by line 120? Did the two distributions perfectly align at 1 $\mu$ m? Or how were the two distributions merged into one?
- I find the description of Section 3.1 and of Fig. 1 over-generalized. As I understand it, you sampled 3 of your carboys at each time point but did not have one system growing during the entire time period. Therefore, I find it overgeneralized to state the bloom peak occurred on Day 9 when we have no continuous record. For example, one of the samples on Day 8 has a higher chlorophyll concentration than one of the samples on Day 9, indicating the bloom of the day 8 sample may have peaked. And for the samples of day 10, we have no indication of when the bloom peak occurred. I suggest rephrasing the results and discussion of the manuscript to consider the lack of continuity, and therefore the ability for larger variation in the time scales of each carboy.

### **Minor Comments:**

- The sentences on lines 53-55 are unclear. Revision is recommended to clarify the point being made.
- Missing references on lines 58-60. What previous laboratory simulations are you referring to?
- Typo on line 116, “ $d_e$ ” should be “ $d_{em}$ ”
- On line 136, are you implying that the concentration of  $Na^+$  remains constant within each experiment, across days, or both?
- Lines 205-206: Typo-- the production of submicron SSA decreased then increased (more or less).
- Lines 231-233: If changes in SML surface tension were responsible for the decrease in SSA concentration and increase in geometric mean diameter, then why is SML surface tension fairly constant after day 7 while the largest changes in SSA occur between days 7 and 9 and again between days 10 and 11?