

Reply to the comments from Anonymous Referee #3

We deeply appreciate Anonymous Referee #3 for the thorough review of our manuscript. Our manuscript has been revised according to the comments and our responses to the comments are as follows. For clarity, the comments are reproduced in blue, authors' responses are in black and changes in the manuscript are in red color text.

Hu et al. describe the wealth of measurements they applied to a multi-part phytoplankton bloom experiment. Specifically, they characterized the POC-DOC cycle across the bloom and through the enrichment and transfer processes among seawater, sea surface microlayer, and sea spray aerosols (separated into submicron and supermicron populations).

I commend the authors in this version of the manuscript. I believe the content and figures are clearer and more specific, and the paper is much stronger than the previous version. The authors have fully addressed the concerns raised during the last round of revisions, and I believe this paper is much stronger. I do recommend for publication after addressing a few more minor edits, see below.

1. The paper would benefit from a final pass through correcting grammatical errors, namely issues with article placement in sentences and use of consistent tenses.

Some examples are below:

- a. Line 241: "As a common..."
- b. Line 242: "...accumulates in the SML..."
- c. Line 243: "...significantly reduces surface..."
- d. Line 252: "...the development of capillary waves..."
- e. Line 254: "...observed in the SML..."
- f. Line 283: "...are pushed upward, form a..."
- g. Line 287: "Compared to the particle size distribution..."
- h. Line 456: "...are "pulse events" in the ocean-atmosphere organic..."
- i. Line 478: "...Chl-a is a driving factor regulating..."
- j. Supplement line 17: "...Copernicus Data Store was checked to determine

whether...”

Author Reply

We have made the corresponding corrections and they are highlighted in the revised manuscript and revised Supplement. Additionally, we have proofread the entire text and corrected grammatical errors, ensuring proper article usage and consistent verb tenses.

2. On line 356, it's stated “Consequently, HULIS1 have greater abundance in the SML and SSA compared to HULIS2 (Fig. 5c).” However, looking at Fig. 5c, it appears that HULIS2 has a greater relative abundance than HULIS1 in the SML. But I agree that $HULIS1 > HULIS2$ is a valid statement for SSA.

Author Reply

We have made the modifications in the revised manuscript.

Page 14, lines 356-357

As a less oxidized organic matter, HULIS 1 exhibits a greater enrichment capacity than HULIS 2 in SSA. Consequently, HULIS1 has a greater abundance in SSA compared to HULIS2 (Fig. 5c).

3. On line 366, it's stated “...however, when sample types differ, significant correlations between them are rarely observed (Fig. 5d).” While I agree with what you're trying to say, of the 14 squares with high correlation (where I'm assuming the white text in the colored box indicates high significance), 4 are between different sample types. I would suggest rephrasing the sentence to better clarify that while most of the high significance correlations occur between samples of the same type, there are 4 cases between samples of different types that exhibit significant correlations.

Author Reply

We have modified the structures of these sentences.

Page 14, lines 363-368

As illustrated in Fig. 5d, although most strong correlations occur between samples of the same type (seawater, SML, submicron SSA, or supermicron SSA), there are four

cases where strong correlations are observed between samples of different types. Among them, the EEM intensities of three dissolved fluorescent components in the SML exhibit significantly positive correlations with the POC concentration in seawater, suggesting that they may be directly derived from seawater POC. Another case is when the strong correlation is observed between HULIS 2 in supermicron SSA and HULIS 2 in seawater, providing indirect evidence that the composition of supermicron SSA and seawater may be closely related.

4. On lines 407-409, the increased percentage of saccharides in SSA is connected to the peak biological activity in the SML. While I think there is a connection, the biological activity and saccharide abundance in the SML doesn't appear to change significantly from day 10 to 14, so I have a hard time believing that's the cause for the shift in SSA saccharide abundance increase on Day 14. I agree they are linked, but I think the statement currently is an over-generalization.

Author Reply

We have provided a more detailed description of this in the revised manuscript.

Page 16, lines 409-412

However, the percentage of saccharides in SSA remains below 10%, with a significant increase only observed on Day 14. This suggests that the abundant saccharides observed in the SML on Day 10 are not immediately and efficiently transferred to SSA. Instead, they likely undergo bacterial modification before manifesting as a significant increase in SSA on Day 14.

5. On lines 435-436 there's a typo with an incomplete reference to a Figure and an accidental paragraph break.

Author Reply

This has been corrected.

Page 17, line 436

Therefore, they exhibit higher EFs in both submicron and supermicron SSA (see Fig. S8).

6. The caption for Figure 6 is incomplete. It's missing a description of panel a, where the current description of a applies to b, and b to c.

Author Reply

We thank the Referee for this remark. We have revised the caption of Figure 6.

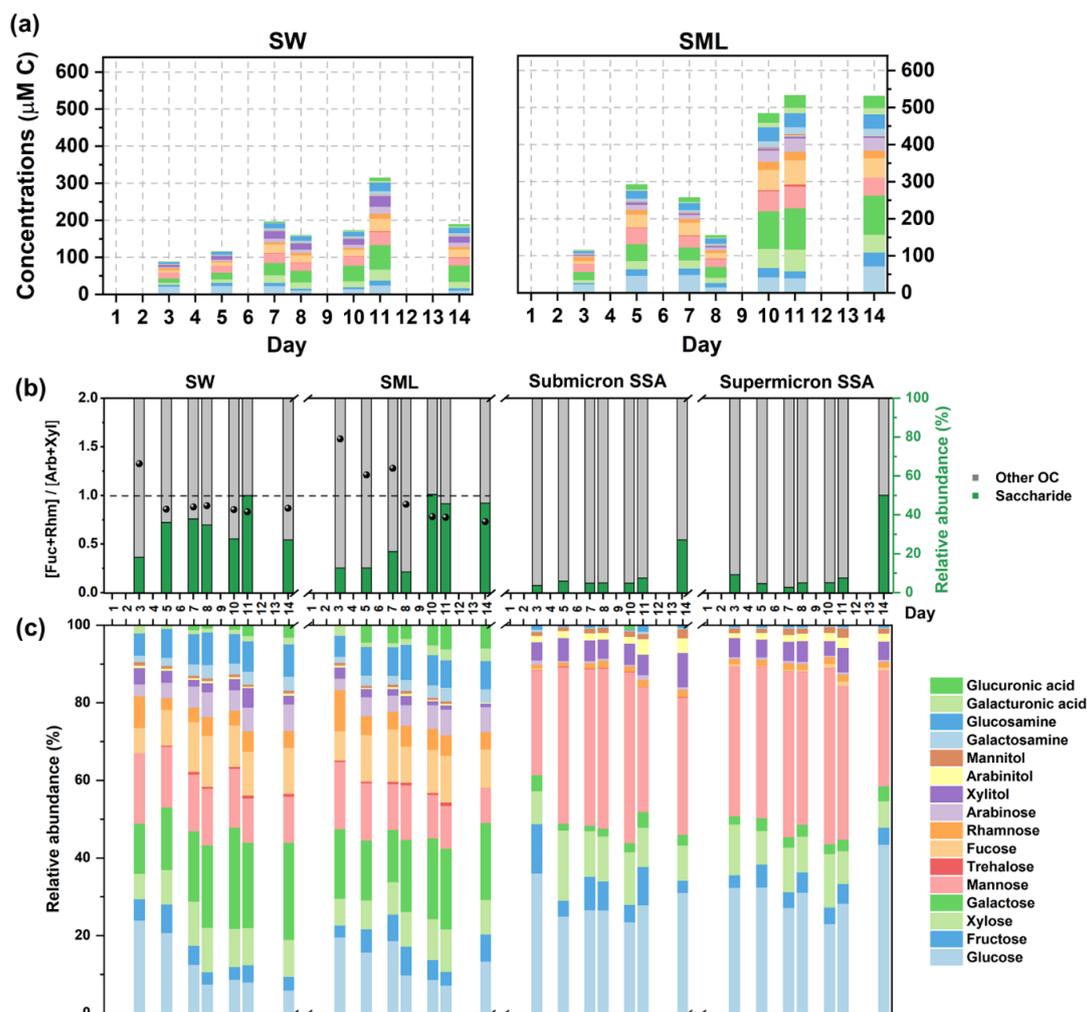


Figure 6. Sea-to-air transfer of saccharides. (a) Time series of saccharides' concentrations in seawater and the SML. (b) Relative abundance of total saccharides in DOC. The bacterial activity is expressed as the ratio of the sum of the concentrations of fucose and rhamnose to the sum of the concentrations of arabinose and xylose (the black scatters). The activity is typically considered higher when the ratio is less than 1. (c) Composition and relative abundance of saccharides.

7. A general comment for section 3.3.3: I agree with all of the information as it is

presented. However, I think it's a bit of an over-generalization to only talk about film drop production impacting submicron SSA and jet drop production impacting supermicron SSA when jet drops produce up to 43% of submicron SSA (see Wang & Deane et al., 2017, PNAS). It's common to generalize with the distinction of film/jet for submicron/supramicron, but I think this paper would be strengthened with a statement acknowledging this generalization and the influence of jet drop production in determining submicron SSA composition.

a. Wang, Xiaofei, Grant B. Deane, Kathryn A. Moore, et al. "The Role of Jet and Film Drops in Controlling the Mixing State of Submicron Sea Spray Aerosol Particles." *Proceedings of the National Academy of Sciences of the United States of America* 114, no. 27 (2017): 6978–83. <https://doi.org/10.1073/pnas.1702420114>.

Author Reply

We have provided a generalized explanation and appropriate references on this point.

Page 17, lines 433-437

Distinguishing film drops from jet drops based on submicron SSA and supermicron SSA is a generalized approach, since submicron SSA also contains a non-negligible contribution from jet drops (~43%) (Wang et al., 2017). However, compared to supermicron SSA (which can be considered entirely composed of jet drops), both film drops and jet drops in submicron SSA are generally inefficient in transferring large particles (Dubitsky et al., 2023; Dubitsky et al., 2024). This may further influence the sea-to-air transfer of polysaccharides.

References

- Dubitsky, L., McRae, O., and Bird, J. C.: Enrichment of Scavenged Particles in Jet Drops Determined by Bubble Size and Particle Position, *Phys. Rev. Lett.*, 130, 054001, 10.1103/PhysRevLett.130.054001, 2023.
- Dubitsky, L., Deane, G. B., Stokes, D. M., and Bird, J. C.: Modeling the Concentration Enhancement and Selectivity of Plastic Particle Transport in Sea Spray Aerosols, *J. Geophys. Res. Oceans*, 129, e2023JC020396, <https://doi.org/10.1029/2023JC020396>, 2024.
- Wang, X., Deane Grant, B., Moore Kathryn, A., Ryder Olivia, S., Stokes, M. D., Beall Charlotte, M., Collins Douglas, B., Santander Mitchell, V., Burrows Susannah, M., Sultana Camille, M., and

Prather Kimberly, A.: The role of jet and film drops in controlling the mixing state of submicron sea spray aerosol particles, Proc. Natl. Acad. Sci. U. S. A., 114, 6978-6983, <https://doi.org/10.1073/pnas.1702420114>, 2017.