

Thölen et al.: Reply to reviewer RC1

We would like to thank RC1 for the careful and constructive comments on our manuscript. We greatly appreciate the detailed comments and suggestions, which have helped us to improve the readability and clarity of the manuscript.

Below, please find your original comments printed in black and our point-by-point responses printed in blue. Line numbers indicated here refer to the preprint manuscript submission.

Please find a list of references used within this answer at the bottom of this document.

RC1: Comment on egusphere-2025-5350, Anonymous Referee #2, 25.02.2026

This manuscript presents the time-course of the concentrations of the colored (CDOM) and fluorescent (FDOM) fractions of dissolved organic matter (DOM) in the sea-surface microlayer (SML) and the underlying water (UWL) during a controlled mesocosms experiment. The impact of a phytoplankton blooms stimulated by nutrient additions and photodegradation processes on the variability of the optical properties of dissolved organic matter in the two layers is assessed.

Overall, this is a well-designed study providing useful insights into the dynamics of colored and fluorescent dissolved organic matter in the SML, specifically regarding its biological production and photochemical decomposition under conditions that allow for isolating both driving forces from the multiple physical and biogeochemical factors involved.

Thank you very much for this positive feedback on our study design.

I find the manuscript to be very thorough, which makes it quite long and occasionally difficult to follow; however, I recognize that this level of detail may be beneficial for readers who are not experts in the optical properties of dissolved organic matter.

Thank you for the feedback on the manuscript length and readability.

Our intention for the manuscript as well as for our subproject in the BASS research group is to address an interdisciplinary readership. We want to show how optical measurements can be applied as proxies for organic matter transformation processes in the sea-surface microlayer. This requires a low-level introduction and method section. Nevertheless, we carefully went through the text and tried to shorten and clarify some of the sentences to ensure better readability and to generally shorten the manuscript without losing depth or information.

Something that strikingly caught my attention is the lack of data on dissolved organic carbon (DOC) concentrations. My recommendation is that, if these were measured, they should be included. If they were not, a brief explanation should be provided.

Thank you for bringing this to our attention. Thanks to your comment we recognize that the connection between CDOM/FDOM and DOC values was missing. DOC was measured during the mesocosm study. DOC data are shown and explained in Bibi et al. (2025a), as well as in the new

preprint Zöbelein et al. (2026). In the supplements of Bibi et al. (2025a) you can find a detailed discussion of an interlab-comparison between the two parties that measured DOC during the mesocosm study. For our manuscript we will show the average DOC data. Using the DOC data we have calculated SUVA₂₅₄ from the absorption at 254 nm and the respective DOC values. We have added two plots to the appendix Figure A2: DOC and SUVA₂₅₄. They are explained in the results Sect. 3.4. When considering the SUVA₂₅₄ results we had to slightly revise the outcome of our second hypothesis as the significantly higher SUVA₂₅₄ in the ULW also supports stronger photodegradation effects in the SML. This is discussed in Sect. 4.2.

Please find a list of detailed comments and recommendations below:

Line 44 – replace 0.4 microm by 0.7 microm

Thank you, we replaced it.

Lines 70 – 74. This is not the right place for these sentences. They should be moved to the last paragraph of the Introduction.

Thank you, we moved the first sentence to the last paragraph of the introduction but rephrased the next sentence so the first hypothesis of the manuscript stays within this paragraph as the sentences before are leading up to it.

Lines 83 – 86. Same as above.

Thanks, we have removed this sentence from the introduction. The same information is already provided in the discussion section and should be sufficient there.

Line 93 – What do you mean with “high resolution” in this context?

Thank you for drawing attention to this phrasing. We have removed the wording “high-resolution” as the resolution of the bio-optical samples was the same as other samples taken during the study.

Nevertheless, it would have been beneficial for the study and the manuscript to have taken bio-optical samples in a higher frequency than the other parameters but unfortunately that was not done because of the limited availability of SML on the mesocosm surface. We had to keep sampling volume to a minimum in order to maintain the SML integrity.

Lines 96 – 98 (Figure 1). Processes excluded in mesocosms studies should be colored differently, e.g. inflow, upwelling, outflow, winds, mixing waves, etc...

That is a good suggestion. We have edited the figure and changed the color of the excluded processes to a lighter blue grey. Additionally, we have printed all the included processes bold and underlined to better differentiate between them. This description was also added to the caption. We would like

to include mixing, because of the flow pumps moving the water column in the mesocosm basin and to exclude waves, which is why we have edited their joint box to two separate boxes within the figure.

Line 101-102. Please specify what is the depth of the outdoor basin (0.8 m). What is the color of the basin? (relevant issue in a study about photodegradation).

Thank you for pointing out that the dimensions could be misleading. We have clarified the dimensions of the basin: [...] The facility contains an 8 m long, 1.5 m wide and 0.8 m deep outdoor basin [...].

We have interpreted your question about the basins color in two ways and hope one of our two responses sufficiently answers the question.

1. The color of the basin itself is a bright concrete grey, which possibly can promote the penetration of sunlight into the basin. We have added this information to the text in the methods and discussion sections.
2. The color of the basin's water changed with the progression of the phytoplankton bloom. Unfortunately, there is no systematic photographic or objective documentation of the color. The information we can provide comes from sporadic pictures and personal memory. The water was greenish, but clear at the beginning of the study and turned milky after the second bloom peak. Bibi et al. (2025a) include a figure of the basin's turbidity measurements. The turbidity increased after the bloom as the coccolithophores shed their coccoliths and turned the water milky.

Lines 104-105. How was constant mixing of the ULW achieved?

That's a good point to add. The mixing was achieved by placing eight flow pumps in the basin. We have added this information to the text.

Line 123. It surprises me that such large water volumes are needed to count **bacterial abundance**.

You are right to wonder about the volume. The colleagues responsible for the bacterial cell count also conducted several other measurements. The large sample volume was required for filtration to accurately characterize microbial community diversity and relative abundances via 16S rRNA amplicon sequencing. They used 400 ml sample volume from SML and ULW each, from which 300 ml was used for filtration, and the rest for enzyme analysis, flow cytometry, and Fluorescence In-Situ Hybridization (FISH). The bacterial cell count method requires 2 ml (specifically 1730 μ l) sample volume.

To clarify this in the text, we have omitted the "because of the large sample volume needed" part of the sentence and just stated the frequency of measurements.

Line 135-136. While storing filtered samples at 4°C is standard practice for FDOM, the duration here extends to several months. Could you specify the exact timeframe? Additionally, were any tests performed to ensure sample integrity after such prolonged storage?

Thank you for raising this point regarding sample storage. After carefully revisiting the measurement documentation we can clarify that all samples were measured within days of being collected, with the exception for the final pair of samples collected on June 16. This pair of SML/ULW samples was measured in September of 2023 together with additional samples collected after the mesocosm study. These additional samples were initially considered to be included in the analysis which led to the original phrasing in the manuscript of being measured “within a few month”. The reason for the delayed measurement was temporary issues with the instrument and another overlapping sampling campaign.

Since it cannot be sufficiently confirmed that the prolonged storage time did not alter the sample composition, we decided to remove the sample pair from 16 June from the data set. According to this we have repeated the analysis, including PARAFAC, to get the accurate data within the given timeframe of 18 May - 15 June. Therefore, the respective figures and tables have been updated (Figures: 2, 4, 5, 6, 7, A1, A2, A3, A4; Tables: 2, 3, 4, 5)

The time frame was indicated in the beginning of the methods section, and the original phrasing was changed accordingly as well to “The samples were stored dark at 4 °C and measured within a few days of the study”.

Line 137. Please, specify the excitation and emission slit widths.

Thank you for drawing attention to this missing information. The Aqualog manual states: [...] the fixed optical geometry of the Aqualog® lends itself to the simple solution above because neither the slit-widths that determine the beam geometry, nor the path-lengths or overlap volume of the absorbance and emission paths are user-adjustable. [...]

The Aqualog manual further specifies a bandpass of 5 nm for the slit width. This information was added to the methods in line 137.

Line 149. No need to include the equation for inner filter correction.

We agree and have removed this equation and its explanation from the text.

Lines 154 and 157. No need to include the equation for Raman normalization.

We agree and have removed this equation and its explanation from the text.

Line 180 – Table 1. IC1, IC2 and IC3 not described yet. Explain meaning in the table caption.

Thank you for pointing that out. We have added their description to the table’s caption (Table 1).

Line 207. Refer to the paper by Bibbi et al. (2025) for nutrient concentrations. A sentence indicating the levels observed and the would help the readers to follow the evolution of the mesocosm.

Adding nutrient information to the results is a good suggestion, thank you for that. We restructured the beginning of the Sect. 3.1, including a new sentence about the nutrient concentrations, and referred to Bibi et al. (2025a).

Line 2014. Which paper of Bibi et al (2025)? 2025a?

Thank you for pointing that out. We missed adding the right reference with the citation program here. This refers to Bibi et al. (2025a). It was changed accordingly.

Line 223. These temperatures seems unrealistic for the Jade Bay. Please comment.

Thank you for drawing attention to the description of the temperature in the mesocosm. We would like to clarify the mesocosm setup: The mesocosm basin is located on-land on the institute's premises. The basin has concrete walls and is quite shallow (0.8 m deep). The incoming sun can reach up until the basins floor and heat up the water column. As there is no exchange with the adjacent Jade Bay during the mesocosm study the water column can warm faster than the bay. The temperature mentioned in this paragraph was measured within the mesocosm basin.

During the time of the mesocosm, we also conducted daily measurements on the Jade Bay. We used the autonomous catamaran HALOBATES (Wurl et al., 2024) to collect additional SML and ULW samples and measure environmental parameters. The location of each deployment of the catamaran was similar every day. Besides other parameters, temperature was measured at 1 m depth. The mean temperature measured by the catamaran HALOBATES rose from about 13.8 °C to 19.5 °C in the Jade Bay in the timeframe of the mesocosm study.

We have also clarified the mesocosms location in the methods: “[...] The on-land facility [...]”. For pictures and a more detailed description of the mesocosm set up we encourage the reader to refer to the paper of Bibi et al. (2025a) at the end of the methods section.

Line 276. It sems more logical to introduce first the variation of the CDOM and FDOM concentrations and later the %FDOM.

Thank you for this suggestion. When revisiting the manuscript, we realized that the information which is provided by Figure 5 is partially redundant to the PARAFAC component time series of Figure 7 in the original submission, now Figure 6. In addition, its interpretation was not integrated into the discussion section and therefore not contributing to the overall narrative. For the sake of clarity and manuscript length, we have removed Figure 5 and its description in the text from the results section.

Line 347. Explain how the sum was calculated. Is just the sum of the Fmax of each component or the sum of the average fluorescence of each component?

Thank you for pointing out that this information could be misleading. The sum refers to the sum of the Fmax of each component. After receiving the comment of the second referee, we decided to remove this sentence here and just include the info in the figure caption. The caption adapted to make this point clearer (Figure 7, now Figure 6).

Line 405 – Figure 8. You may calculate the derivative of Chla in the panels on the right.

That's a good suggestion. After revisiting the figure, we have decided to shorten it, by not including the REPIX index and the C1/a312 index, as the slopes of these indices are neither strongly supporting nor contradicting the hypotheses. We adapted Figure A2 in the appendix to include the timeseries of REPIX and C1/a312 as well as the beforementioned SUVA₂₅₄. We have updated former Figure 8, now 7, to include a plot with the deviation of chlorophyll-a. We decided against showing it in every plot on the derivation side as it's very noisy during the bloom phase even after smoothing advances. We also followed up on the new indications provided in the text in the results and discussion section.

Lines 536-537. a₂₅₄ should not change substantially because natural UV of this wavelength does not arrive to the Earth Surface so it does not directly decompose chromophores absorbing at this wavelength.

We agree that solar radiation at 254 nm does not reach the Earth's surface and therefore does not directly induce photodegradation of chromophores absorbing at this wavelength. However, changes in a₂₅₄ do not necessarily require direct irradiation at 254 nm. CDOM absorption arises from a continuum of electronically coupled chromophores rather than independent absorbers, as shown by Del Vecchio and Blough (2004). In such systems, photochemical alteration at longer wavelengths (e.g., UV-A and visible) can induce changes across the full absorption spectrum.

Accordingly, the observed changes in a₂₅₄ in our study are interpreted as the result of indirect photochemical transformation processes affecting the overall DOM pool, rather than direct photodegradation at 254 nm.

Line 542-543. This sentence is repeated in the previous paragraph. It could be erased.

We agree with this observation. We have added the information of this sentence to its appearance in the previous paragraph to refrain from doubling information but keeping the indication that the increase in SR values suggests that photodegradation outweighed microbial alteration as a sink.

Lines 555-561. This paragraph about a paper in preparation could be omitted.

Thank you, we agree that a paper in preparation should be omitted, when the manuscript was first written the submission of Zöbelein et al. (2026) was very close, so we decided to add its info to the

text. In the meantime, the paper Zöbelein et al. (2026) was also posted as a preprint at EGU sphere. As its results underline the findings in our paper, we would much like to include it.

Line 574. The conclusions may be drastically reduced. Lines 576–581 and 591–594 can be omitted.

Thank you for this suggestion. Those sentences were removed from the conclusions.

Additional references

- Bibi, R., Ribas-Ribas, M., Jaeger, L., Lehnert, C., Gassen, L., Cortés-Espinoza, E. F., Wollschläger, J., Thölen, C., Waska, H., Zöbelein, J., Brinkhoff, T., Athale, I., Röttgers, R., Novak, M., Engel, A., Barthelmeß, T., Karnatz, J., Reinthaler, T., Spriahailo, D., ... Wurl, O. (2025). Biogeochemical dynamics of the sea-surface microlayer in a multidisciplinary mesocosm study. *Biogeosciences*, 22(23), 7563–7589. <https://doi.org/10.5194/bg-22-7563-2025>
- Del Vecchio, R., & Blough, N. V. (2004). On the Origin of the Optical Properties of Humic Substances. *Environmental Science & Technology*, 38(14), 3885–3891. <https://doi.org/10.1021/es049912h>
- Wurl, O., Gassen, L., Badewien, T. H., Braun, A., Emig, S., Holthausen, L. A., Lehnert, C., Meyerjürgens, J., & Ribas-Ribas, M. (2024). HALOBATES: An autonomous surface vehicle for high-resolution mapping of the sea-surface microlayer and near-surface layer on essential climate variables. *Journal of Atmospheric and Oceanic Technology*. <https://doi.org/10.1175/JTECH-D-24-0021.1>
- Zöbelein, J., Sawle, S., Friedrichs, G., Ribas-Ribas, M., Lehnert, C., Paetz, K., Pflaum, M., & Waska, H. (2026). Buoyancy and polarity driven accumulation of dissolved organic matter in the sea surface microlayer during a phytoplankton bloom. *Biogeochemistry: Organic Biogeochemistry*. <https://doi.org/10.5194/egusphere-2025-6563>