

We thank the **co-editor-in-chief** for the constructive comments and valuable feedback on our manuscript. In the following table, we address each comment individually and describe the changes and improvements made to the manuscript.

| Comment | Response |
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| L33: I would like to suggest to replace 'hotspot' with 'reactor'. Your main conclusion is that the SML behaves like a biogeochemical reactor. | As suggested, we have replaced 'hotspot' with 'reactor' in the revised manuscript (L34). |
| L198-L203: Inorganic nutrients analysis: Please add the detection limits and the analytical errors. | We have added the detection limits and analytical error for the inorganic nutrient measurements in the revised manuscript (L204–L206). |
| L214: surfactant film abundances: Would it be possible to give an estimate of the error associated with these measurements? | Thank you for pointing this out. The uncertainty of the surface coverage (<i>sc</i>) are now included in the experimental section of the revised manuscript (L225 L-227). We have also updated the relevant formula for <i>sc</i> in the supplementary material (L128-128). |
| L373-374: I am a little bit confused about the apparently very short spring time in northern Germany. It says that 18 May highlights early spring conditions and 12 June reflects summer conditions implying that there was only a month from early spring to summer. Please rephrase | Meteorological spring in northern Germany spans March–May; therefore, 18 May reflects late spring conditions , while 12 June represents summer conditions. This has been rephrased in the revised manuscript (L376-L377). This reflects the meteorological seasons in Germany. |
| L407: The concentration range given for phosphate in the SML is wrong; see Figure 4d. Please correct. | We thank you for pointing out this oversight. The phosphate concentration in the SML has been corrected in the revised manuscript (L409). |
| Figures 4d and 4e: I would like to suggest to change the y-axis in Fig 4d to | Figures 4d and 4e have been revised to use a log-scale on the y-axis, in line with the suggestion. |

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| <p>a log scale since the unusual high phosphate conc. in the SML after the bloom do mask the variability of phosphate at low conc. Moreover, is difficult for the reader to justify the increasing N:P ratios in the ULW during the first week of the study (Fig. 4e): Decreasing nitrate (Fig.4a) and the apparently constant phosphate conc (Fig. 4d) would result in decreasing N:P ratios during the first week. But I think the increasing N:P ratios were resulting from a relatively more pronounced decrease in phosphate conc in the ULW; but this cannot be inferred from Fig. 4d.</p> | |
| <p>Figures 7b-g: Please apply the same time axis as in Figure 7a and indicate the bloom as you did in Figure 7a.</p> | <p>The time axis has been revised to align Figure 7a with Figures 7b-g. Please note that for some dates, substrate utilization data were not available (Figures 7b-g); therefore, the corresponding gaps in the x-axis represent days without data.</p> <p>We have added the bloom phase indication in Figure 7b-g in alignment with Figure 7a.</p> |
| <p>L640-641: I would like to suggest to remove 'wet' or replace it with 'dry' because wet deposition is mentioned as 'precipitation' later in the same sentence.</p> | <p>The sentence has been rephrased in the revised manuscript (L644-L645).</p> |
| <p>L681-682: E. hux growth during the pre-bloom phase: Assuming that the parameters shown in Fig. 5c indicate E. hux abundances, then I do not see an</p> | <p>In Fig. 5c, the 5–10 μm size class, which represents coccolithophores such as <i>Emiliania huxleyi</i>, shows elevated integrated volume concentrations ($\mu\text{L L}^{-1}$) from 18 May to 26 May. This contrasts with the abundance of <i>Cylindrotheca</i></p> |

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| <p>increase during the pre-bloom phase which, in turn, implies that <i>E. hux</i> was not growing in the pre-bloom phase. In my point of view, this seems to be in contrast to your statement.</p> | <p><i>closterium</i> (Fig. 5d), whose cell numbers (cells L⁻¹) only began to increase after nutrient additions and during the bloom phase, with a marked rise from 29 May onwards. The temporal dynamics of chlorophyll c concentrations (Fig. 5b), a marker pigment for haptophytes including coccolithophores (<i>E. huxleyi</i>), also coincide with the 5–10 µm size class patterns. In addition, the 3–4 µm size class in Fig. 5c likely represents detached coccoliths from <i>E. huxleyi</i>. These were relatively low during the pre-bloom phase while <i>E. huxleyi</i> populations were still growing, but increased once <i>E. huxleyi</i> growth began to decline, reflecting enhanced coccolith detachment.</p> |
| <p>L763: I guess 'matric' should read 'matrix'</p> | <p>This has been corrected to 'matrix' in the revised manuscript (L770).</p> |

Anonymous Referee #1

We sincerely thank the reviewer for their thorough and thoughtful evaluation of our manuscript and for recognizing its scientific merit, novelty, and overall quality. We appreciate the constructive suggestions, which have helped us further improve the clarity and precision of our manuscript.

| Comment | Response |
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| I note only a single minor technical adjustment, one formula would benefit from an explicit definition of a parameter to eliminate any residual ambiguity. This is a straightforward revision that the authors can very easily accommodate. | We thank the reviewer for noting this oversight. In the revised manuscript, we have provided an explicit formula for the enrichment factor as $EF = C_{SML} / C_{ULW}$, where C_{SML} and C_{ULW} are the concentrations of the variable in the SML and ULW, respectively (line 298-299). All parameters are now clearly defined to avoid ambiguity. |
| Line 21 – For clarity, please specify where the mesocosm experiment was carried out so that the experimental setting is clear from the abstract alone, without readers needing to consult the full paper. | We have specified the location of the mesocosm experiment, the Center for Marine Sensor Technology (ZfMarS), Institute of Chemistry and Biology of the Marine Environment (ICBM), Wilhelmshaven, Germany, to clarify the experimental setting directly in the abstract. |
| Line 185 – It is unfortunate that analyses for DOC, PON, TDN, PN, and pigments could not be carried out on the all SML samples; however, this statement contradicts the information in line 223 and again in the Results section (line 490), where it is reported that both SML and ULW samples were analyzed for DOC and TDN (see line 495). This discrepancy is confusing for the reader and should be checked and corrected. | We have corrected the inconsistency regarding DOC and TDN analyses between Line 185 and Lines 223, 490–495. The revised text now consistently states which parameters were analyzed for SML and ULW samples. Specifically, DOC and TDN were analyzed for both SML and ULW samples, whereas POC and PN were analyzed only for ULW samples. This change ensures clarity and consistency across the Materials and Methods and Results sections. Please note that the TDN dataset is currently undergoing re-evaluation. Any updates resulting from this process will be incorporated into the revised final |

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| | manuscript but will not affect the overall conclusions of the study. |
| Line 413 – Minor suggestion: Although readers in this field will understand the context, I recommend adding parentheses around the sum of the N-nutrients so that it is mathematically explicit. | We have revised the expression of the sum of N-nutrients, $((\text{NO}_3^- + \text{NO}_2^-): \text{PO}_4^{3-})$, adding parentheses for mathematical clarity. |

Anonymous Referee #2

We thank the reviewer very much for their constructive comments and valuable feedback on our manuscript. In the following table, we address each reviewer's comment individually and describe the changes and improvements made to the manuscript.

| Comment | Response |
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| <p>The identification of <i>Emiliania huxleyi</i> as the main bloom former needs stronger evidence. The authors should provide morphological data from SEM analysis confirming the presence of <i>E. huxleyi</i> cells and characteristic coccoliths.</p> | <p>Morphological confirmation:</p> <p>We now provide SEM images showing coccoliths characteristic of <i>Emiliania huxleyi</i> and intact cells of <i>Cylindrotheca Closterium</i>, as shown in Figure R1. FlowCam images collected after removal of the size filter (<10 µm) also reveal numerous small cells consistent with <i>E. huxleyi</i>. Together, these images confirm the presence of these species within the experimental system and have been included in the Supplementary Information (Fig. S6) in the revised manuscript.</p> <div style="display: flex; justify-content: space-around;"> <div style="text-align: center;"> <p>(a)</p> </div> <div style="text-align: center;"> <p>(b)</p> <p>SEM HV: 10.0 kV WD: 8.37 mm Det: SE 10 µm VEGA3 TESCAN View field: 49.8 µm Date/midday: 10/04/23 © SENCKENBERG am Meer SEM MAO: 7.25 kx WD: 8.37 mm Det: SE 10 µm VEGA3 TESCAN Date/midday: 10/04/23 © SENCKENBERG am Meer</p> </div> <div style="text-align: center;"> <p>(c)</p> <p>SEM HV: 10.0 kV WD: 8.37 mm Det: SE 10 µm VEGA3 TESCAN View field: 49.8 µm Date/midday: 10/04/23 © SENCKENBERG am Meer SEM MAO: 7.25 kx WD: 8.37 mm Det: SE 10 µm VEGA3 TESCAN Date/midday: 10/04/23 © SENCKENBERG am Meer</p> </div> </div> <p>Figure R1. Bloom-forming species observed during the mesocosm experiment. (a) FlowCam images of <i>Cylindrotheca closterium</i> (numbers indicate image identifiers). (b) SEM image of <i>C.</i></p> |

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| | <p>closterium. (c) SEM image of coccoliths belonging to <i>Emiliania huxleyi</i>.</p> |
| <p>Additionally, molecular validation would be helpful. DNA analysis, such as qPCR with primers targeting the genomes of <i>E. huxleyi</i> and <i>Cylindrotheca Closterium</i>, would confirm species abundance.</p> | <p>We acknowledge the value of DNA-based methods (e.g., qPCR) for species-specific quantification. However, such analyses were beyond the scope of this mesocosm experiment, and no remaining sample material is available for retrospective molecular analysis. We agree on the importance of such approaches and will consider integrating them into similar experiments in the future.</p> <p>In the revised manuscript, we (i) clarify the FlowCam acquisition settings and their limitations in the Methods section (line 255), while presenting the associated findings in the Results (Species Identification) section (line 446-451) and (ii) include the SEM and FlowCam images (Supplementary Fig. S6). We think these additions and clarifications strengthen our identification of <i>E. huxleyi</i> as the main bloom former in this study.</p> |
| <p>Lastly, why wasn't the FlowCam useful for detecting <i>E. huxleyi</i> as it was for diatoms? Other studies have used it for such analysis before.</p> | <p>Initially, FlowCam imaging was performed with a >10 µm size filter to focus on larger diatoms, as the experimental design aimed to trigger a diatom bloom following nutrient addition. This prevented the early detection of smaller cells such as <i>E. huxleyi</i>. After an increase in turbidity suggested the presence of other taxa, the filter was removed, revealing abundant small cells later confirmed via SEM as <i>E. huxleyi</i>. However, these FlowCam data only cover the latter half of the experiment, limiting their use for full temporal bloom reconstruction.</p> |
| <p>It is most important to accurately describe the phytoplankton community, as the organic matter composition in the SML and ULW is expected to differ significantly between algal species, however the current</p> | <p>We have strengthened our species identification by including SEM and FlowCam images confirming the presence of <i>E. huxleyi</i> and <i>C. closterium</i> (provided as Supplementary Fig. S6 in the revised manuscript). We have also clarified the limitations of our initial FlowCam settings in revised manuscript.</p> |

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| <p>species identification presented seems unconvincing.</p> | |
| <p>The statement that "coccoliths shed by <i>E. huxleyi</i> during the final stages of the bloom significantly increased water column turbidity and light scattering" lacks supporting evidence for a coccolithophore bloom, as mentioned earlier. Using 3–4 µm size bins in FlowCam as a proxy for <i>E. huxleyi</i> coccoliths is highly speculative, especially without the demonstration of co-occurrence of <i>E. huxleyi</i> cells. This observed particle fraction might most likely be composed of other pico-eukaryotes or cyanobacteria. Authors should provide convincing evidence for the presence of these coccoliths, or otherwise, should temper their somewhat speculative conclusions that phytoplankton blooms contribute to climate-relevant feedback mechanisms through increased turbidity and albedo effects caused by coccolith shedding from <i>E. huxleyi</i> blooms.</p> | <p>We now provide SEM images as evidence for coccolithophore presence, showing both intact <i>Emiliania huxleyi</i> cells and detached coccoliths during the late phase of the bloom (Supplementary Fig. S6).</p> <p>Contribution to turbidity and optical properties:</p> <p>In our mesocosm experiment, we observed a measurable increase in water column turbidity and albedo during the late bloom phase, coinciding with SEM-confirmed coccolith shedding and an increase in the LISST 3–4 µm particle size fraction (compare Fig. 2b and Fig. 5c). These observations are consistent with the previous established knowledge showing that coccolithophores produce calcite plates (coccoliths) contribute to increased turbidity and a whitish appearance of the water column (Holligan et al, 1983; Beaufort et al., 2008; Perrot et al., 2018) and coccolithophore blooms increase backscattering and enhance albedo due to their high refractive index and strong light-scattering properties (Balch et al. 1999, 2005, 2011; Tyrrell et al. 1999; Frouin & Lacobellis, 2002; Gordon et al., 2009; Tyrrell & Merico, 2004; Fournier & Neukermans, 2017) on regional to global scales. We acknowledge that other particles (e.g., <i>C. closterium</i> cells and detrital material) may also have contributed to turbidity; our SEM evidence and particle size distribution data indicate that coccoliths played a substantial role in the observed turbidity change.</p> <p>Revised climate-relevant conclusions:</p> <p>While we observed increased turbidity and albedo (Fig. 2b and 3c) coinciding with coccolith presence (Fig. 5c), we agree that attributing global-scale climate feedback mechanisms solely to coccolith shedding in this mesocosm experiment exceeds the scope</p> |

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| | <p>of our study. We have therefore revised the discussion (line 648-650; line 696-699) and conclusion paragraph (line 809-812) to emphasize that our findings represent local experimental conditions and are consistent with previously documented optical effects of coccolithophores.</p> |
| <p>The bacterial abundance determination through SYBR staining and flow cytometry raises concerns too: the non-selective nature of the SYBR green dye may potentially lead to overestimation of cell concentration. In addition, I couldn't find a description or a chart of the FCM gating plots. This gating strategy should be clearly described, presenting the positive control bacterial cells compared to the bacterial cell counts from the environmental samples analyzed.</p> | <p>We understand the reviewers' concern about accurate cell counts; however, the method we used for determining bacterial abundance is standard in microbial ecology (Marie et al., 1999; Brussaard et al., 2010). To ensure accurate quantification, the flow rate was calibrated using fluorescent bead standards. Though SYBR Green can also bind to non-DNA particles, leading to inaccurate results, especially in complex samples like soil. This nonspecific binding can cause overestimation of DNA or reduced accuracy in quantification. However, in our case, samples were filtered through a 5 μm filter to remove most larger particles and non-bacterial debris, thereby minimizing the likelihood of overestimation.</p> |
| <p>Could the author also explain why cells were not counted through CFU plating or molecular analysis (e.g., qPCR)? It is highly recommended to add such analysis to the manuscript as a validation. Otherwise, the limitations of the SYBR green labeling method should be discussed, and FCM bacterial counts should be termed</p> | <p>While CFU counts are valuable for estimating viable bacteria, they rely on the ability of cells to grow on a given culture medium. In marine environmental samples, typically only 1–10% of bacteria are culturable under laboratory conditions, meaning CFU counts would significantly underestimate total bacterial abundance and therefore would not be directly comparable to flow cytometry results. Similarly, qPCR can help quantify specific bacterial taxa; however, it is not optimal for estimating total bacterial abundance in diverse marine samples. This is because marine bacteria vary greatly in the number of ribosomal RNA operon copies per genome, which can bias total abundance estimates.</p> |

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| <p>"bacterial-like particle" counts for accuracy.</p> | <p>Flow cytometry with SYBR Green staining is a widely used, established method in marine microbial ecology for estimating total bacterial abundance, and results are directly comparable to other studies using similar methods. In light of the reviewer's suggestion, we will adopt the term "bacterial-like particles" when referring to our flow cytometry counts. However, this terminology is not used in marine microbial ecology.</p> |
| <p>Another important point that raises questions about the validity of the bacterial count method is the atypical bacterial growth (Figure 7a). Values are expected to be higher by orders of magnitude, as observed in other <i>E. huxleyi</i> microbiome studies.</p> | <p>The primary objective of the mesocosm study was to obtain a comprehensive understanding of the synergistic and antagonistic interactions among multifaceted biogeochemical processes in the SML and ULW during the development and decline of an induced phytoplankton bloom. Accordingly, our study was not designed as a microbiome study of <i>Emiliania huxleyi</i>, although this species was blooming during the experiment. Variability in bacterial cell counts likely reflects the influence of multiple environmental and biogeochemical factors beyond <i>E. huxleyi</i> abundance, which may have contributed to the observed lower cell numbers.</p> |
| <p>Overall, the manuscript would benefit from a description of what has been previously published versus the novel contributions of this study.</p> | <p>We have revised the <i>Discussion</i> (line 629-640; line 704-711) to explicitly compare our findings with previously published studies, most of which were conducted in field settings and focused on individual aspects of the SML, rather than broader interactions such as SML-ULW coupling and the interplay between physical, chemical, and biological processes. We now clearly highlight the novel contributions of our study, which include presenting the first comprehensive and integrated measurements of biological (phytoplankton biomass and community composition, bacterial abundance and metabolic profiles), chemical (nutrients and surfactants), and physical (turbidity and solar irradiance) parameters from paired SML and ULW samples. In addition, our study captures the coupling of complex, multifaceted biogeochemical processes between the SML and ULW over the full</p> |

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| | <p>course of a phytoplankton bloom succession within a controlled mesocosm setting.</p> <p>Please note that Referee #1 considered the novelty of our work clear and highlighted that our approach and findings represent a valuable addition to the existing literature.</p> |
| Minor Comment. Several figure captions do not accurately describe the content of the figure panels. | <p>We have carefully and thoroughly reviewed and revised all figure captions and ensured that each accurately and clearly describes the content of its corresponding panels, including all symbols, abbreviations, and units where applicable. We believe these changes will improve clarity and consistency across all figures.</p> |

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