

General changes by the authors:

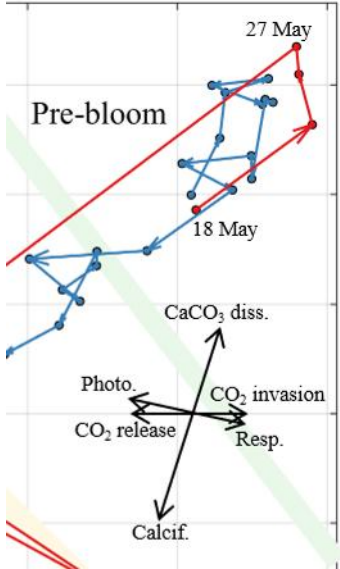
We sincerely thank the Editor and Reviewer for their time, thorough reading, and feedback where needed. Comments have helped us to improve the clarity and overall quality of this manuscript.

Reviewer 1:

No.	Comment	Response
1	Page 1, Line 29: Abstract section: “Spearman $\rho = -0.75$, $p = 0.024$ ” – The correlation is mentioned but the sample size ($n=10$?) should be stated for transparency.	We have corrected the sentence in the revised manuscript: Page 1, Line 31: (Spearman $\rho = -0.76$, $n = 10$, $p = 0.024$)
2	Page 2, Lines 56-57: Introduction section: The sentence “ <i>Emiliana huxleyi</i> consume HCO_3^- to form coccolith and produces acidic polysaccharides associated with dissolved oranic matter. From diatoms, <i>Cylindrotheca closterium</i> , release organic material related to carboxylate-rich extracellular polymeric substances” contains multiple errors. “oranic” → “organic”, “related” → “related”. The sentence structure is fragmented and grammatically incorrect.	We have corrected the sentence in a new paragraph and included the specific organics: Page 2, Lines 69-73: Coccolithophores (<i>Emiliana huxleyi</i>) calcify by using Calcium (Ca^{2+}) and DIC, mainly precipitating HCO_3^- to build up calcite coccoliths. This calcification is mediated by acidic, Ca^{2+} -binding polysaccharides (De Jong et al., 1979), whose carboxyl and sulphate groups are proton-active once released to seawater and can thus contribute to the organic matter (Passow, 2002). As another example, diatoms (<i>Cylindrotheca closterium</i>) release predominantly polysaccharide-rich exudates containing uronic acids and sulphate ester groups (Staats et al., 1999).
3	Page 3, Line 77: Introduction section: “under ssurfactant-enriched” → under surfactant-enriched	This grammatical error was corrected as follows: Page 3. Line 87: The SML is consistently enriched in surface-active organic material.
4	Page 4, Lines 100-110: Method section: The mesocosm setup description is adequate, but several details are missing. What was the: 1) light:dark cycle?	1) Light:dark cycle: We did not explore the light:dark cycle because the lack of SML data (due to the limited volume that could be sampled) resulted in insufficient resolution.

2) How was temperature controlled/monitored? 3) What was the actual nutrient concentration added (μM)?	The mesocosm was exposed to natural ambient light in an outdoor facility with a light-transmissive roof. Regarding discrete samples, even though ULW was collected daily and alternated between dark (~0300 UTC) and light (~1300 UTC) conditions, the SML sample volume for each day was limited because it had to be distributed among the BASS subprojects participating in this study. Therefore, we received SML samples only every third day, reducing exploration of the data for light:dark cycles. We have added clearer details of the mesocosm setup as follows: Page 5, Lines 136-137: Discrete samples for the parameters DIC, TA, and OA were collected from both layers as part of the multidisciplinary BASS collaboration under alternating natural-light (~1300 hrs UTC) and dark (~0300 hrs UTC) conditions. 2) Temperature controlled/monitored: We have changed the text as follows: Page 4, Lines 120-123: The temperature from the SML was recorded (Rauch et al., 2026) using two temperature microsenors (UNISENSE, Denmark; tip diameter: 180 μm). The SML salinity was measured directly from the discrete sample using a conductivity meter (Cond 340i, WTW, Germany). For the ULW at 40 cm, temperature and salinity were measured using a CTD48Mc (Sea&Sun Technology, Germany) that continuously monitored (Bibi et al., 2025). 3) Nutrient concentration added: We have added the following details on nutrients addition. All concentrations are reported in $\mu\text{mol L}^{-1}$:
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		Page 4, Line 126-129: To induce a bloom, inorganic nutrients were added on 26 May (Nitrogen (N): 0 $\mu\text{mol L}^{-1}$, Phosphorus (P): 1.2 $\mu\text{mol L}^{-1}$, Silicate (Si): 19.8 $\mu\text{mol L}^{-1}$), 30 May (N: 10 $\mu\text{mol L}^{-1}$, P: 0.6 $\mu\text{mol L}^{-1}$, Si: 10 $\mu\text{mol L}^{-1}$) and 01 June (N: 5 $\mu\text{mol L}^{-1}$, P: 0.3 $\mu\text{mol L}^{-1}$, Si: 0 $\mu\text{mol L}^{-1}$) to achieve a N:P:Si ratio of 16.7:1:16.7 (Bibi et al. 2025).
5	Page 5, Lines 125-130: Method section: The glass plate technique sampling speed ($\sim 5 \text{ cm s}^{-1}$) is reported, but the film thickness achieved is not stated. This is important for comparing with other studies.	In Rauch et al. (2026), we reported a thickness of $\sim 937 \pm 369 \mu\text{m}$ as a diffusion and thermal boundary layer across the study. We added the following: Page 5 Line 138-142: The SML samples were collected using a pre-cleaned glass plate (Harvey & Burzel, 1972) that was immersed through the SML, moved away from the perturbed immersing area, and withdrawn vertically. Manually controlled $\sim$at a rate of 5 to 6 cm s^{-1} as suggested by Carlson (1982); at such speed the glass plate can collect a microlayer of $52 \pm 1.8 \mu\text{m}$ thickness (mean $\pm 95 \%$ CI, and multiple dips were combined to reach the required sample volume.
6	Page 7, Lines 215-225: Method section: The bloom phase definition based on chlorophyll-a is mentioned, but actual Chl-a values are not provided in the main text, including representative values for each phase.	The referred statement was moved to chapter 2.1 “Mesocosm study set-up, induced bloom and bloom phases definition” and reads now as follows: Page 4, Lines 126-130: We defined the three bloom phases based on chlorophyll a (Chl <i>a</i>) concentrations according to Bibi et al. (2025): pre-bloom phase with Chl <i>a</i> concentrations averaged/below $2.2 \mu\text{g L}^{-1}$ (18 to 26 May), the bloom phase with average Chl <i>a</i> concentration of $7.3 \mu\text{g L}^{-1}$ (27 May to 05 June), and post-bloom with Chl <i>a</i> concentration below $1.8 \mu\text{g L}^{-1}$ (05 to 16 June)
7	Page 8, Line 238: Results section: “CaCO₃ dissolution appears to have been the dominant process in the SML”	We interpreted Line 238 (in the original manuscript) as follows:

	– This interpretation is not strongly supported by the vectors in Figure 1c, which show mixed signals. Please revise or provide additional evidence.	Looking at the start to end of the pre-bloom phase, from 18 May to 27 May, the resultant vector is aligned with CaCO_3 dissolution.  Page 10, Line 301-303: In the SML, NDIC remained at high concentrations between 2025.07 and 2180.39 $\mu\text{mol kg}^{-1}$ (Error! Reference source not found.a). In comparison, NTA rose continuously from 2286.05 to 2434.92 $\mu\text{mol kg}^{-1}$ (Error! Reference source not found.b). This was shown from the start to end of the pre-bloom phase, from 18 May to 27 May; the resultant NDIC vs NTA vector is aligned more with CaCO_3 dissolution (Error! Reference source not found.c).
8	Page 10, Figure 2: Results section: The pie charts showing OA/TA fractions are informative, but the exact values (8.1% vs 3.1%) differ slightly from the abstract (8.4% vs 3.1%). Please clarify.	Both the pie chart and the text showing the OA contribution to TA were unified to 8.1%. Page 1, Line 29-29: OA was persistently enriched in the SML relative to the ULW (enrichment factor: $\text{EF} > 1$ on nearly all sampling days) and contributed, on average, 8.1% of the TA, compared with a low 3.1% in the ULW Page 11, Line 348-349:

		in our study, SML OA accounted for a larger fraction of TA (8.1%) than ULW (3.1%).
9	Page 12, Figure 4: Results section: The confidence interval for the slope should be reported in the figure caption.	We added the confidence interval and the interval of the slope: Page 14, Figure 4 caption, Line 396-397: (Slope: $b = -0.053 \Delta\text{pH}$ per OA EF unit, 95% CI = [-0.1029, -0.0028]; $n = 10$).
10	Page 13-15, Lines 349-415: Results section: The discussion section is largely descriptive, restating the results rather than exploring mechanistic explanations for OA enrichment and pH modulation. Key mechanistic questions are unaddressed: (1) Why is OA enriched in the SML during bloom phase transitions (e.g., phytoplankton exudation vs. microbial processing vs. physical accumulation)? (2) What specific functional groups of dissolved organic matter (DOM) drive OA in the SML?	We agree that the previous version of the discussion was too descriptive. The revised discussion now opens with a mechanistic framing paragraph connecting the OA EF- ΔpH correlation to the acid-base system behavior of proton-active organic compounds and then addresses both questions in dedicated paragraphs. (1) Three mechanisms account for OA enrichment at bloom transitions: Phytoplankton exudation Microbial transformation Active surface accumulation (2) Functional groups driving OA in the SML We acknowledge that direct molecular speciation was not performed and explicitly identify this limitation as a priority for future work. Summary of changes to section 4 (Discussion): (i) Opening paragraph: mechanistic framing of the OA EF- ΔpH relationship. (ii) Second paragraph: NDIC-NTA biological process sequence as context. (iii) Paragraphs 3-5: one paragraph each for phytoplankton exudation, microbial transformation, and surface-active accumulation. (iv) Paragraph 6: acid-base system chemistry of the functional groups found and quantification of pH impact.

		(v) Paragraph 7: literature comparison of OA concentrations (retained from v4). (vi) Paragraph 8: caveats and priorities for future work (retained and refined). (vii) Paragraph 9: methodological framework and three-layer model (retained and refined).
11	Page 14, Lines 385-395: The comparison with literature values is useful, but the authors should address why their ULW OA values (40-100 $\mu\text{mol kg}^{-1}$) exceed typical coastal values – could this be an artifact of the mesocosm system or the measurement method?	We acknowledge that studies reporting OA in this field remain relatively limited, which constrains direct comparisons with the literature. The elevated ULW OA values observed in our study likely reflect the specific conditions of the mesocosm experiment. In particular, the system experienced an intense bloom event (see also Bibi et al. 2025; Rauch et al., 2026), which likely enhanced the production and accumulation of organic matter and, consequently, OA beyond levels typically reported for coastal environments. This effect may have been further amplified by the confined nature of the mesocosm, where organic matter can accumulate without the dilution processes characteristic of open-ocean settings. We also consider the possibility that OA may be overestimated under mesocosm conditions due to the presence of organic compounds that are not fully titratable but still contribute to the measured signal. However, regarding potential methodological artifacts, we took great care during method development and quality assurance as described in the section on methodologies. This included the use of internal reference materials, procedural blanks, and rigorous validation steps to ensure the reliability of our measurements. Therefore, we are confident that the reported values are not analytical artifacts but rather reflect the combined influence of bloom dynamics, confinement effects, and the system's specific chemical composition.