General changes by the authors:

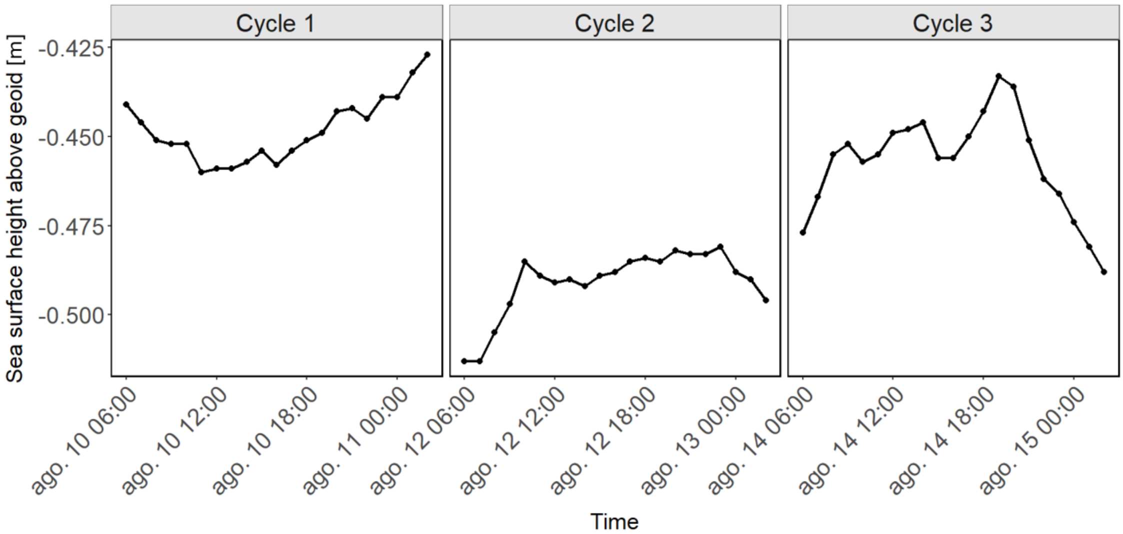
We have revised the manuscript by reorganising the Methods and Results sections for improved clarity, refining the description of the sampling strategy and measurement procedures, and strengthening the discussion of the physical and biogeochemical drivers of diel variability. Additional supporting figures (tidal height and $p\text{CO}_2$ time series) have been included, and the title, Abstract, and Conclusions have been updated accordingly. All reviewer comments have been fully addressed.

Reviewer 1:

Comment	Response
Title: While the authors infer some diel variability in the inorganic marine carbon system in the surface microlayer from their pH, T, salinity, measurements, as the title implies, the authors note that the main implication of the results pertains to the fact that neglecting nocturnal fluxes leads to a systematic bias in CO_2 fluxes across the air-sea interface. They further note that the differences in these fluxes is driven by the wind. With this in mind, the current title doesn't capture that main implication, and the authors might consider a title change.	We appreciate the reviewer's thoughtful suggestion regarding the title and the recommendation to reflect better the implications of diel variability for air–sea CO_2 exchange. While our study primarily focuses on the diel dynamics of the inorganic carbon system in the sea-surface microlayer, we agree that highlighting their influence on CO_2 flux estimates would improve clarity and better communicate the broader relevance of our findings. In response to this comment, we have modified the title accordingly. The revised title now reads: **"Diel Variability Affects the Inorganic Carbon System in the Sea-Surface Microlayer and Influences Air–Sea CO_2 Flux Estimates."** We think this revised title maintains the study's original focus while explicitly acknowledging the implications for CO_2 exchange, thereby addressing the reviewer's concern.
line 108-109 (in the track changes version) the authors note that precipitation averaged 1.4 mm over the first half of August. Is that value 1.4 mm per day? It seems there should be a time component to this average rate.	The reported precipitation value corresponds to the mean daily rainfall during the days preceding the sampling campaign, not the total accumulation or the average over the whole month. To clarify this, we have revised the sentence in the text (Line 104 in the track changes file).
lines 129-130, the authors note that "the sampling location was treated as a single point, since spatial variations within the region were minimal." Spatial variations in what parameter(s)? And what does "minimal" mean? Can the authors elaborate	The statement refers to the spatial scale covered by the sampling platform. According to GPS positions recorded during the campaign, the catamaran remained within a very small area, with a maximum horizontal displacement of approximately 90 m across all cycles. At this spatial scale, no meaningful horizontal gradients

on what constitutes minimal variation in the parameters of interest?	in surface temperature, salinity, or pH_{T25} are expected; therefore, the site can be reliably treated as a single representative point. The text has been revised accordingly to clarify this point (Lines 138-142).
line 362, the sentence "This response reflects both the temperature dependence of carbonate speciation." "Both" implies that the authors will list 2 things that carbonate speciation is dependent on. I think they left a word out here.	The original sentence aimed to describe the combined influence of temperature-driven physical processes on pH_{T25}, but the wording was indeed unclear. This section has now been rewritten (Lines 353-361), and the term "both" no longer appears. The revised text explicitly distinguishes between the lack of a direct temperature effect on carbonate speciation (because pH is expressed as pH_{T25}) and the indirect influence of temperature-dependent physical processes such as evaporation and vertical mixing.
lines 402-403, the authors note that the observed variability follows a diurnal cycle rather than a semi-diurnal cycle. This statement is used as evidence for the absence of a tidal explanation for changes in physicochemical parameters, and this may be true. It definitely appears true that the T data and portions of the salinity (cycle 3) and pH (Cycle 3) data show diurnal variations (Fig. 3), but, for example, the Cycle 2 Salinity data and Cycle 1 pH data show patterns that may be semi-diurnal, and the Cycle 1 salinity data show an unclear trend. A plot of the tidal cycle on the figure and/or a statistical test of tide height versus these parameters or of the periodicity of cyclic patterns would provide the reader more confidence of the lack of tidal influence.	We fully agree that evaluating the potential tidal influence is important. As suggested, we examined the local sea level time series during the sampling period using Copernicus MEDSEA reanalysis data (product MEDSEA_MULTIYEAR_PHY_006_004), since in-situ tidal records from the Hydrographic Institute of the Republic of Croatia (HHI) were not accessible for the period examined. Copernicus MEDSEA has been extensively validated for sea-level applications in the Mediterranean and provides a scientifically robust alternative for characterising tidal dynamics in microtidal environments. The resulting tidal signal is minimal, with a peak-to-peak range of approximately ~0.1 m, consistent with the microtidal character of the central Adriatic. According to the HHI, average tidal ranges based on long-term sea level measurements have been detected as follows: 0.22 m in Dubrovnik, 0.23 m in Split, 0.25 m in Zadar, 0.30 m in Bakar, and 0.47 m in Rovinj (https://www.hhi.hr/en/about-us/projects/adriatic-sea-level). Such a low-amplitude variation would appear nearly flat relative to the diel variability observed in temperature, salinity, and pH. It, therefore, would provide limited explanatory value if superimposed on Fig. 3. Moreover, we found no coherent semi-diurnal (~12 h) pattern in the physicochemical variables. The diel variability is not phase-locked across cycles: Cycle 1 displays no consistent sub-daily structure; Cycle 2 salinity shows a sub-daily fluctuation not reproduced in temperature or pH; and Cycle 3 shows a clear diurnal signal. This lack of stable periodicity and phase alignment is incompatible with a tidal origin, as tidal signals

	maintain highly stable frequency and phase over consecutive days. For this reason, overlaying the tidal height on Fig. 3 would not improve the interpretation of the data: the tidal curve would appear nearly flat relative to the magnitude of diel variability, and the two signals do not align in phase or periodicity. Nonetheless, in response to the reviewer's helpful suggestion, we will include the tidal time series for the sampling period as supplementary material (Figure S1), allowing readers to independently evaluate the tidal context of our measurements (Line 150).



Figures S1. Sea surface height above geoid (m) during the three sampling cycles, obtained from the Copernicus Marine Service reanalysis product MEDSEA_MULTIYEAR_PHY_006_004 (https://data.marine.copernicus.eu/, last accessed 15 November 2025).

Reviewer 2:

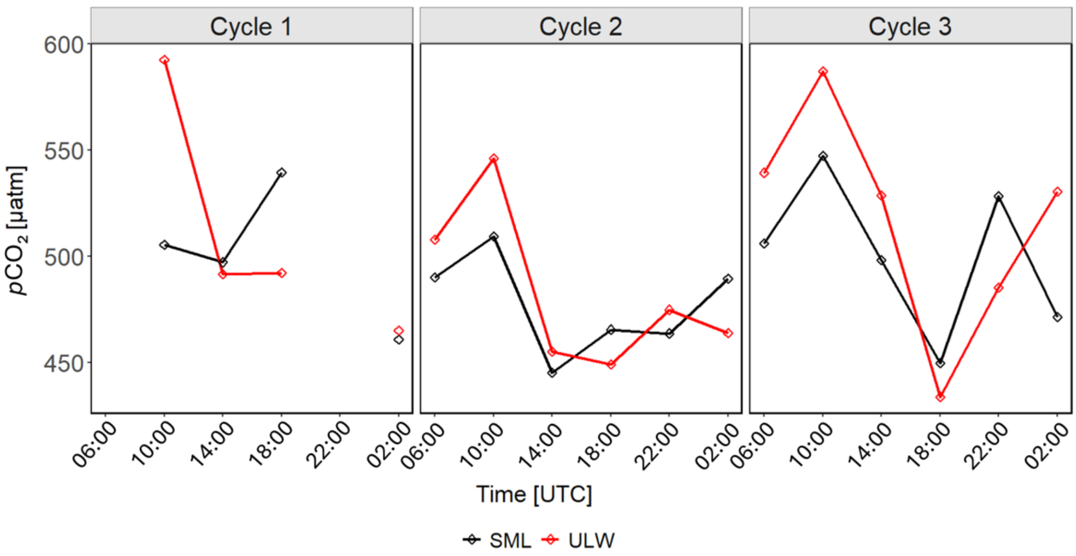
Comment	Response
In this revised version, the authors have greatly improved several parts of the manuscript, adding methodological information that now allows a better understanding of the work conducted and the results presented. The description of the study area and sampling strategy is now appropriate, and the methodological section has been improved. However, it would still benefit from further reorganization, with a clearer and more ordered description of the equipment, sampling strategy, and analytical methods. The results section would benefit from some synthesis, a careful check of the subsection	We thank the reviewer for the positive assessment of the improvements introduced in this revised version and for the constructive suggestions provided. Following the recommendations, we have further reorganised the Materials and Methods section to present the sampling equipment, sampling strategy, and analytical procedures in a clearer and more ordered structure. We have also revised the Results section to improve synthesis, ensure consistency in subsection titles, and reinforce the distinction between the presentation of results and their interpretation. Regarding the suggestion to merge Figures 5 and 6 into a single figure, we fully acknowledge the intention to facilitate

titles, and a clearer distinction between results and discussion. I would also suggest combining Figures 5 and 6 into a single figure to facilitate easier comparison.	comparison. However, we respectfully consider that keeping them separate improves clarity and readability. Each figure supports a distinct key finding in an associated paragraph, and merging them would result in a visually dense figure and a less direct link between the figures and their corresponding interpretations. For this reason, and to preserve the alignment between the figure structure and the narrative flow, we propose retaining the current figure organization.
Line 108: "the Zodiac" correct?	We have changed the text accordingly.
Line 111. This is still not clear. Is there a single sensor package that measures the SML continuously and can be switched to the ULW? Ore there are two sensor packages?	The SML and ULW were monitored in parallel using two independent sensor packag**es** mounted on the S^3 system, each dedicated to one water layer. Thus, temperature, conductivity/salinity, and pH were measured simultaneously in both the SML (collected via rotating glass discs) and the ULW (drawn from 1 m depth). We have now clarified this explicitly in the revised manuscript (Lines 114-116, in the track changes file).
Line 113. Please move the reference to the table after the sensor package (or sensors); it does not refer to the automated collector.	We have changed the text accordingly.
Line 115. How many discrete samples per deployment?	We have now clarified the sampling design. For each sampling event, one discrete bottle was collected for DIC and one for TA at each sampled depth, resulting in paired DIC–TA determinations for each diel observation. This information has been added to the Methods section.
Line 116. approaching the catamaran with the support vessel Zodiac?	We have changed the text accordingly.
Lines 118-120. this is sampling strategy, I suggest to move this at the end of the section together with the other part addressing this aspect	We agree with the reviewer and have moved this text to the end of Subsection 2.1, following the description of the sensors, measurements, and analytical procedures. The content has been integrated into a single unified paragraph that describes the overall sampling strategy.
Lines 125-130. This part describes the sampling strategy: I suggest to move it at the end of the section, after the description of the sampling and measurements,	See previous comment.
Line 131. Subsamples of what samples?	We have clarified the origin of the subsamples by specifying that they were derived from the SML and ULW water collected by the S^3 system (Line 126).
Line 208. Please provide some numbers	We now provide the specific measurement precisions that constrain the uncertainties in σt

	and pCO_2 (temperature ±0.1 °C, salinity ±0.2 %, pH ±0.005; see Table 1).
Line 288. "Table S2" please move at the end of the sentence, this table provides the results of the statistical tests	We have changed the text accordingly (Line 281).
Line 298. "The large variability…" discussion	The sentence has been revised to avoid interpretative wording and to remain fully descriptive within the Results section (Line 290).
Line 304. Title 3.4 "Biogeochemical processes variability across diel cycles. Here the same parameters were presented, with a focus on higher frequency data. I suggest to modify the title, as it is a bit misleading	We agree that the original title could imply a broader biogeochemical scope than what is actually presented in this subsection. To improve clarity and better reflect the focus on high-frequency variability of the measured physicochemical parameters, we have revised the title to: "High-resolution variability of physicochemical parameters across diel cycles."
L. 340: The discussion of gas fluxes and their drivers lacks presentation of pCO_2 data. Only the difference in pCO_2 between the two layers is shown. Given the daily variations in temperature (and often in pH), a significant diurnal cycle is expected. These data could be added to Figure 7 and presented in the Results section.	Thank you for pointing out the absence of pCO_2 data in the presentation of CO_2 fluxes. We agree that showing the diel behaviour of pCO_2 can provide helpful context for interpreting the fluxes. However, adding additional panels to Figure 7 would introduce substantial redundancy with the existing presentation of pH_{T25}, ΔpCO_2, and the fluxes themselves. To address your suggestion while keeping the Results section concise, we have now included the full SML and ULW pCO_2 time series in the Supplementary Material (Figure S2), and we refer to this figure in the Results section (Lines 343). This provides readers with direct access to the diel variability in pCO_2 without altering the structure of the main figures.
L. 355–365: The results are presented in a somewhat confusing way and lack clarity. I suggest separating the discussion of the drivers of night/day variability from that of vertical variability. Clearly explain first the effect of temperature, and then that of salinity on the other parameters.	To improve clarity, we have revised the structure of this paragraph so that the discussion explicitly separates the two levels of variability addressed in it: (i) diurnal (day-night) variability driven by changes in solar radiation, wind forcing, and evaporation, and (ii) vertical variability between the SML and the ULW. First, we have explicitly focused on the meteorological factors that drive diurnal variability and their influence on temperature, salinity, and pH_{T25}. Next, we have added a concise explanation highlighting the vertical differences between the two layers, emphasizing that the SML responds more strongly to surface forcing than the ULW (Lines 351-360).

L. 360. "This response reflects both the temperature dependence of carbonate speciation, (Zeebe & Wolf-Gladrow, 2001)" if pH is normalized at 25°C there is no temperature dependence on carbonate species composition and pH	We agree that pH_{T25} does not retain the temperature dependence of carbonate speciation. To address this, we reformulated the sentence to reflect the correct mechanism: the variability in pH_{T25} is linked to temperature-driven physical processes (evaporation and vertical mixing) that modify DIC and TA, rather than to a direct temperature effect on carbonate chemistry (Lines 355-360).
L.375: "The calculated mean temperature values correspond to the Middle Adriatic Surface Water mass (Table S1)." I cannot find these values, please add them to the text to allow an easier comparison. Something is missing in the sentence, are the values higher, lower?	The reference to Table S1 was previously misplaced, which may have made it challenging to locate the corresponding mean temperature values. We have now rewritten the sentence to clearly indicate that the mean SML and ULW temperatures across the three cycles (reported in Table S1) fall within the expected range for the Middle Adriatic Surface Water mass. The revised text (Lines 369-371) also retains the following sentence, which provides previously published summertime temperature values for comparison.
L. 414: "Cantoni et al 2012" To observe the day/night variability you need high frequence measurements, that are not presented in this paper. You could look at the following work, https://doi.org/10.1016/j.rsma.2021.101770 from Ragazzola et al 2021 It is very coastal but from a Mediterranean region	We appreciate the reviewer's observation regarding the use of Cantoni *et al.* (2012). Since this reference does not explicitly address diel variability, we have replaced it with Ragazzola et al. (2021), which more directly documents day–night changes in pCO_2 and pH in a Mediterranean coastal setting. The sentence has been updated accordingly (Line 406).
L. 415–424: The discussion of the carbonate system remains confusing. pCO_2 is not normalized to 25 °C, and therefore it also depends on temperature — higher pCO_2 values are expected during the day for this reason. pCO_2 calculated at 25 °C could be used to discuss processes other than temperature. Please revise the discussion accordingly.	We agree that the previous text did not clearly distinguish the thermal effect on *in situ* pCO_2. In the revised manuscript, we now explicitly state that pCO_2 was not normalised to 25 °C, and therefore its daytime increase is primarily explained by the temperature dependence of CO_2 solubility (Takahashi et al., 1993; Weiss, 1974). We also clarify that this physical effect operates alongside biological and mixing processes that may further modulate the diurnal signal. The paragraph has been rewritten accordingly to provide a more precise and more accurate interpretation of the observed pCO_2 variability (Lines 406-413).
L. 435–460: Please improve the discussion by also considering pCO_2 variability. Usually, FCO_2 is calculated taking into account wind speed over the 24-hour cycle (as such data are usually available), and these speeds are then applied to pCO_2 values measured only during the day. Your dataset allows you to estimate the potential bias introduced by this approximation. You could also discuss	We have expanded the Discussion (Lines 440-452) to explicitly address the diel variability of pCO_2 and its potential drivers. Using our SML pCO_2 measurements, we now report the magnitude of day–night differences across the three cycles (e.g., ~53 µatm in Cycle 1 and <2 µatm in Cycles 2 and 3). We also evaluate the processes likely responsible for this variability: temperature-driven solubility

whether the main driver of day/night $p\mathrm{CO_2}$ variability is temperature variation, biological processes, or buoyancy fluxes, among others. These could be very interesting insights.	changes and intermittent wind- and buoyancy-driven mixing appear to dominate, while a persistent biological day–night signal is not evident in our dataset. In addition, we quantify the slight bias introduced when using daytime-only $p\mathrm{CO_2}$ values (e.g., +13 µatm in Cycle 1) and contrast it with the much larger bias resulting from neglecting nighttime CO_2 fluxes (33–50%). These additions clarify the relative importance of physical and biogeochemical processes in shaping diel CO_2 exchange and directly address the recommendation.
Finally, please update the Abstract and Conclusions accordingly.	To ensure internal coherence with the expanded Discussion, both the Abstract and the Conclusions have been updated to keep aligned with the new wording without adding further length or detail to the section.



Figures S2. Time series of $p\mathrm{CO_2}$ in the SML and ULW across the three diel cycles.