

## Reply to Reviewer #1

We sincerely thank Dr. Brian Butterworth for his positive feedback and constructive comments, which have significantly improved the quality of the manuscript. All coauthors have worked together to revise the manuscript accordingly. In the text below, the reviewer's comments are in **black**, our reply is in **blue**, and the revision of the manuscript is in **orange**.

The manuscript entitled “Mechanisms of air-sea CO<sub>2</sub> exchange in the central Baltic Sea” by Yuanxu Dong et al. describes the results of a field campaign to measure CO<sub>2</sub> fluxes in the Baltic Sea, with a specific focus on additional physical processes (beyond wind speed) influencing gas exchange. The EC measurements appear to have been collected with the proper technical methods. The manuscript is well written and the figures clearly present the results. While there are certain built-in limitations from the relatively short period of data collection (e.g., limited range of SA and wind speeds), the study provides a worthwhile contribution to the field by testing key advancements to CO<sub>2</sub> flux estimation. The multivariate (SA, Hs, ustar) parameterization for k660 provides a straightforward path for incorporating additional physics and provides a useful starting point for future studies. I recommend the paper for publication after some minor comments are addressed.

Thank you for your thoughtful comments and for recognizing the value of our study.

### Comments:

Line 26: What does “40% stronger seasonal cycle of CO<sub>2</sub> flux” mean precisely? Larger difference between annual high and low values? Larger magnitude fluxes overall?

It is the larger difference between the high and the low values. In detail, the summer uptake is enlarged and the outgassing in winter is also enhanced due to the increase of  $K_{660}$  in all seasons. We added this extra information to help the reader understand.

When applied to climatological forcing, it yields a 40% stronger seasonal cycle (greater oceanic uptake during summer and enhanced outgassing during winter) of CO<sub>2</sub> flux in the Baltic Sea than obtained with the conventional  $U_{10N}$ -based parameterization.

Line 54: “affected by many factors near-surface processes” is redundant.

Thank you. We removed this redundant information.

Equation 1 highlights the central role of  $K_{660}$  as the kinetic forcing parameter in air-sea  $\text{CO}_2$  exchange.  $K_{660}$  is directly driven by near-surface turbulence (Garbe et al., 2014).

Line 72-73: awkward comma splice

Thank you. We revised this sentence by changing a comma to a bracket to improve the clearness.

Wave breaking is strongly impacted by the fetch (defined as the distance over which wind acts on the water surface), because limited fetch suppresses wave breaking and bubble generation.

Line 108: change “mole density” to “molar density”

Yes. Revised.

where  $\rho$  is the mean molar density of dry air

Line 125: It’s not necessary, but it could help future groups, if you mention that this configuration (i.e., 3/8” ID, 33.2 lpm flow rate) results in turbulent flow within the tube.

Good point. Thank you.

Air was drawn from the port-side inlet through a ~10-m Teflon tube (3/8" inner diameter) at a stable flow rate of  $33.2 \pm 0.3 \text{ L min}^{-1}$ , which results in turbulent flow within the tube.

Line 129: The Edson et al. (1998) paper did not have a procedure to correct for misalignment between the anemometer and the motion sensors. Miller et al. (2008; <https://doi.org/10.1175/2008jtecho547.1>) added an alignment transformation matrix.

Did you use this?

Thank you for pointing out this. This is a key procedure for the motion correction. Based on the statement from the Miller et al. (2008) (see the screenshot below), for small offset angles, we could simply add the offset angles to the Euler angles. Therefore, we generally include an *alpha* and a *beta* angle to account for the rotation of the sonic relative to the IMU. I checked that the angle offsets between the sonic and the IMU are zero both horizontally and vertically during the CenBASE cruise.

$$\mathbf{u} = \mathbf{T}_{ep}\mathbf{M}_{pa}\mathbf{u}_a + \mathbf{T}_{ep}\left(\int \ddot{\mathbf{x}}_p dt + \boldsymbol{\Omega}_p \times \mathbf{r}_p\right) + \mathbf{v}_{\text{ship}}. \quad (4)$$

In E98, the motion sensor and anemometer were mounted together so their coordinate axes were coaligned and  $\mathbf{M}_{pa} = \mathbf{I}$ , where  $\mathbf{I}$  is the identity matrix. For the small offset angles we measured (less than  $7^\circ$ , Table 2), we found that the matrix multiplication  $\mathbf{T}_{ea} = \mathbf{T}_{ep}\mathbf{M}_{pa}$  in Eq. (4) was closely approximated by simply adding the offset angles to the calculated Euler angles, that is,  $\mathbf{T}_{ep}\mathbf{M}_{pa} = [\psi_{ep}][\theta_{ep}][\varphi_{ep}]\mathbf{M}_{pa} \approx [\psi_{ep}][\theta_{ep} + (\bar{\theta}_{ep} - \bar{\theta}_{ea})][\varphi_{ep} + (\bar{\varphi}_{ep} - \bar{\varphi}_{ea})]$ .

Here we added the Miller et al. (2008) to show that we have accounted for this issue.

Data processing and quality control procedures followed those described in Dong et al. (2021).

Briefly, motion corrections were applied to the wind (Edson et al., 1998; Miller et al., 2008)

Miller, S. D., Hristov, T. S., Edson, J. B., & Friehe, C. A. (2008). Platform motion effects on measurements of turbulence and air-sea exchange over the open ocean. *Journal of Atmospheric and Oceanic Technology*, 25(9), 1683–1694. <https://doi.org/10.1175/2008JTECHO547.1>

Line 152: This is confusing: “according to the open ocean EC cruise tracks (see Yang et al., 2022)”.

Do you simply mean you extracted ERA5 wave parameters according to the same method as Yang et al. (2022)? Based on a couple sentences down it appears you perform an analysis using previous EC cruises? If so, it hasn’t been introduced yet.

That’s why the wording on Line 152 is confusing.

Sorry for the confusion. We agree that the previous open-ocean EC data should first be introduced.

Here we added this information and rephrased the sentence:

In addition, EC air-sea CO<sub>2</sub> flux observations from previous open-ocean cruises (Yang et al., 2022) are also used to comparison with the CenBASE results. Wave parameters were extracted from the ERA5 analysis wave product according to these open-ocean EC cruise tracks (see Yang et al., 2022) and the CenBASE cruise.

Line 186: Add “coming” or “being obtained” (or similar) before “from” in “leading to most valid EC measurements from outside this period”

Yes. Thank you, Added.

leading to most valid EC measurements being obtained from outside this period

Line 197: The sentence that starts with “This supports...” needs work. As it’s written, it needs and object after “supports” (e.g., “idea”). But the sentence is wordy. Here’s a suggested modification: “This suggests that the COARE model remains applicable in fetch-limited marine environments when wave information is included, despite being developed primarily from open-ocean observations.”

Thank you. We agree to your suggestion. The sentence is revised as:

This suggests that the COARE model remains applicable in fetch-limited marine environments when wave information is included, despite being developed primarily from open-ocean observations (Edson et al., 2013).

Line 218: DT Experiment is summarized in Appendix 1 (not 2).

Good spot. Thank you.

*K<sub>660</sub>* from the DT experiment is summarized in Appendix A1.

Line 231/233: “parameterization” and “parameterisation” are used in the same caption (and throughout the manuscript). Choose one for consistency.

Nice point. We revised all the “parameterization” and “parameterized” into “parameterisation” and “parameterised”, respectively.

Line 247: As this is currently written it sounds only theoretical. Might be worth citing Yang et al. (2022) here, as this was empirically found (see the last sentence of the article).

Yes. We agree. The Yang et al. (2022) is added:

This is unsurprising since the chemical enhancement (Cole & Caraco, 1998; Fairall et al., 2022; Yang et al., 2022) and likely buoyancy flux sustain CO<sub>2</sub> transfer at low winds (McGillis et al., 2004; Wanninkhof et al., 2009).

Line 266: It’s not clear to me what you did here. “Following this separation framework” sounds like you ran the same machine-learning analysis as Yang et al. (2024) to get the coefficients in Eq. 3. Did you do that? If not, where do they come from? I don’t see that exact equation in Yang et al. (2024).

Sorry for the confusion. The equation is directly taken from Equation in Yang et al. (2024). The equation is originally expressed in  $\text{m s}^{-1} * 360000$  (equal to  $\text{cm hr}^{-1}$ ) in Yang et al. (2024):

$$K_{660} = K_{i660} + K_{b660} = 360000 * (1.52 * 10^{-4} * u_* + 2.90 * 10^{-5} * u_* H_s)$$

and here I expressed it as  $\text{cm hr}^{-1}$  directly with  $360000 * (1.52 * 10^{-4}) = 55$  and  $360000 * (2.90 * 10^{-5}) = 10$ .

To avoid the confusion, we rephrase this sentence to make it clear.

Following this separation framework and the open ocean EC data analysis, Yang et al. (2024) express the  $K_{660}$  as (Fig. 4B, black line):

$$K_{660} = K_{i660} + K_{b660} = 55u_* + 10u_*H_s \quad (3)$$

Line 273: I suggest adding a little more explanation of what these modeled values represent. It was not clear to me initially whether the modeled values in this paragraph were the model applied to the Yang 2024 open ocean data or whether they were the model applied to Baltic cruise data that had similar wind speeds to the open ocean data, but with  $H_s$  for the Baltic Sea (which you do show later). It became clear later, but would help to clarify it here. How did you select for similar wind speeds? Was it simply all open ocean data points with wind speeds below 12 m/s? Or did you match wind speed distributions?

Thank you. We agree to this valuable suggestion. They were the modelled values with the Baltic cruise wind speed data. Equation 3 provides a parameterisation of  $K_{660}$  applicable to the open ocean conditions. Here we aim to show what the mean  $K_{660}$  values would be if there was a cruise in the open ocean that mirrored the windspeed conditions during the Baltic Sea cruise? By comparing the modelled open-ocean  $K_{b60}$  value with the observed  $K_{660}$  value in the Baltic Sea, we can estimate the suppression.

To clarify this, we added more information and rephrased the sentences:

The observed EC  $K_{660}$  during CenBASE was on average  $14.9 \text{ cm hr}^{-1}$ . To compare this value with open-ocean conditions at equivalent wind speeds, we apply the wind-speed observations from the CenBASE cruise (i.e., wind speed values shown in Fig. 4A) to Equation 3 to estimate open-ocean  $K_{660}$ , yielding average values of  $K_{i660} = 15.1 \text{ cm hr}^{-1}$ ,  $K_{b660} = 7.0 \text{ cm hr}^{-1}$ , and  $K_{660} = 22.1 \text{ cm hr}^{-1}$ . This means that the observed  $K_{660}$  during CenBASE was 33% ( $7.2 \text{ cm hr}^{-1}$ ) lower than the open-ocean  $K_{660}$  estimate.

Line 274: “yields on average values” to “yields average values”

Thank you. Revised.

apply the wind-speed observations from the CenBASE cruise (i.e., wind speed values shown in Fig. 4A) to Equation 3 to estimate open-ocean  $K_{660}$ , yielding average values of  $K_{i660} = 15.1 \text{ cm hr}^{-1}$ ,

Line 290: Appears to be missing “and the CenBASE cruise track”

Yes. Revised.

The data are extracted from ERA5 according to the EC cruise tracks (Yang et al., 2022) and the CenBASE cruise track.

Figure 5B: If I understood correctly, the solid black and blue are total  $k_{660}$  from the Yang et al. (2024) equation and the dashed black and red isolate only the  $k_b$  component. If so, the notation in the legend does not clearly convey that. Specifically, it’s unclear what “ $H_s$ ” means. It’s not immediately intuitive that it means total  $k_{660}$ . Also, since the solid blue and the dashed red lines are related in the same way as the solid and dashed black lines, I’d suggest using the same color for them.”

Good idea. Thank you. We have revised the legend and the color to make their representation clearer:

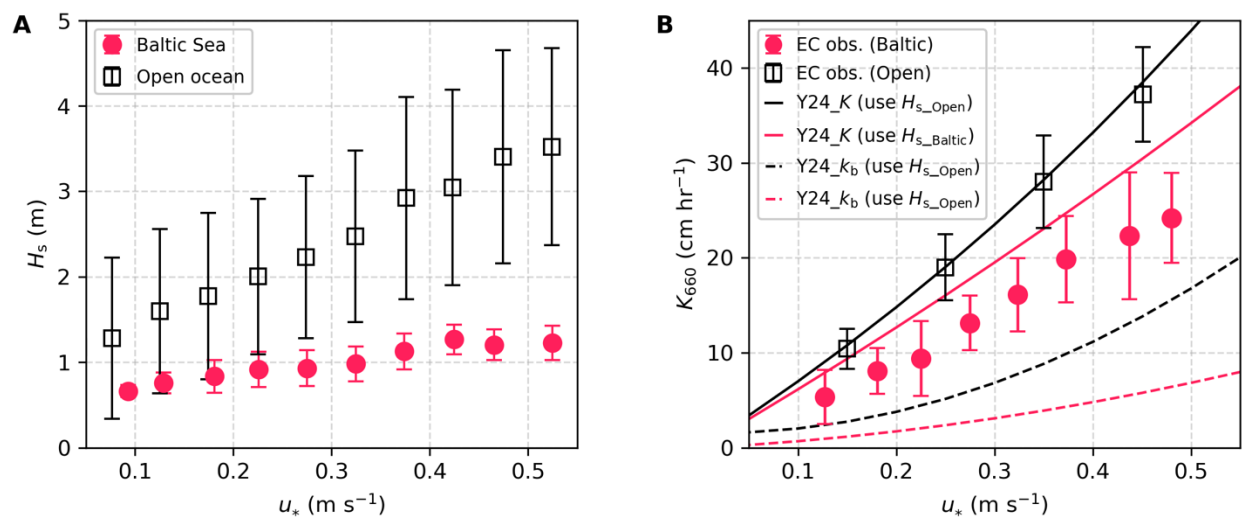


Figure 5: Comparison of significant wave height ( $H_s$ ) and  $K_{660\_CO2}$  between the Baltic Sea and the open ocean. A:  $H_s$  in the Baltic Sea during CenBASE (red dots) and in the open ocean (black squares),

with error bars representing  $\pm 1$  STD. The data are extracted from ERA5 according to the EC cruise tracks (Yang et al., 2022) and the CenBASE cruise track. **B**:  $K_{660\_CO_2}$  observations in the Baltic Sea during CenBASE (red dots, the same as the red dots in Fig. 4B) and in the open ocean (black squares, Yang et al., 2022). The black and red solid lines correspond to the parameterised total  $CO_2$  transfer velocity (i.e.,  $K_{660\_CO_2}$  from Equation 3; Yang et al., 2024) using the open ocean and the Baltic Sea  $H_s$ , respectively. The black and red dashed lines denote the parameterised bubble-mediated transfer component ( $K_{b660}$ ; Equation 3) using the open ocean and the Baltic Sea  $H_s$ , respectively.

Table 1: It would also help to be more explicit in the table what is modeled and what is measured. For example, adding “EC” somewhere on the “Baltic Sea (CenBASE)” line and “model” somewhere on the “open ocean” line. Separately, “Gas Transfer velocity (cm hr-1)” heading is over the wrong column. Maybe it could be placed over the k columns?

Nice suggestions. Revised.

		$K_{i660}$ (cm hr <sup>-1</sup> )	$K_{b660}$ (cm hr <sup>-1</sup> )	$K_{660}$ (cm hr <sup>-1</sup> )	Uncertainty (cm hr <sup>-1</sup> )	
Open ocean (model)		15.1	7.0	22.1	$\pm 5.5$ ( $\pm 25\%$ )	
Impact factors (model)	Fetch	$u_*$	+1.5 (+10%)	+0.7 (+10%)	+2.2 (+10%)	$\pm 0.6$ ( $\pm 25\%$ )
		$H_s$	0	-4.0 (-57%)	-4.0 (-18%)	$\pm 1.6$ ( $\pm 40\%$ )
	Surfactants	Unsure	Unsure	-5.4 (-25%)	$\pm 5.8$ ( $\pm 107\%$ )	
Baltic Sea (CenBASE, EC)		-	-	14.9 (-33%)	-	

Figure 6: Yang21 isn't cited in the caption, but the rest of the studies are.

Good spot. Thank you. Cited.

Red squares represent values derived from an EC-based  $CO_2$  transfer velocity study (Yang et al., 2021).

Line 356: Because the SA suppression is inferred, there is no way to test this. But do you think that the two  $sf$  corrections are independent? Would you expect an additional interaction term (e.g.,  $coeff * SA * ustar$ ) to modulate the resulting  $sf$ ? Could that explain the different slopes from different studies in Fig 6b (i.e., they have different SA concentrations and therefore have different  $sf$  vs  $ustar$  relationships)?

This is an intriguing question. To our knowledge, no dedicated studies have examined whether the two corrections are independent. We adopt the  $SA$  dependence of  $sf$  from Pereira et al. (2018), while the  $u_*$ -dependent correction is extrapolated from the expected variation in  $SA$  concentration with  $u_*$ . We attribute the variation of  $SA$  concentration with  $u_*$  to the mixing level: stronger wind stress enhances upper-ocean mixing, thereby reducing  $SA$  enrichment in the sea surface microlayer. This represents a physical process by which surface dynamics modulate surfactant enrichment at the sea surface. In contrast, the dependence of  $sf$  on  $SA$  concentration likely arises from the damping of surface turbulence and the reduction in effective surface area available for gas exchange. This reflects a process whereby enriched surfactants influence surface turbulence and gas exchange. It is plausible that the effects of mixing on  $SA$  enrichment and the modulation of surface turbulence by  $SA$  are not mutually independent. Nevertheless, our correction in Equation 5 implicitly assumes independence between these two processes. To account for this potential interaction, we explicitly address this concern in the uncertainty analysis section:

Furthermore, it is worth noting that the two corrections in Equation 5 (i.e., the  $SA$ - $sf$  correction and the  $u_*$ - $SA$  correction) are implicitly assumed to be independent. However, potential interactions between  $u_*$ -dependent  $SA$  variation and the  $SA$  influence on  $sf$  may introduce additional uncertainty into Equation 5.

Line 374: It's not clear how the 20% uncertainty was calculated. The word "assign" sounds like it was a rough estimate (which would be fine). But if it was more quantitative than that, it would be worth elaborating.

Yes, it is too simple to use "assign" for this uncertainty analysis. Yang et al. (2024) show that the  $R^2$  of the fit is  $\sim 0.75$ , which means a  $\sim 25\%$  variation of the observed  $K$  is still not able to be explained by the parameterisation. Here, we revised the text and adopt this number as an uncertainty of the parameterisation.

Yang et al. (2024) reported that the  $R^2$  for the fit (i.e., Equation 3) is  $\sim 0.75$ , indicating that  $\sim 25\%$  of the variance in the observed  $K$  remains unexplained by the parameterisation. We therefore assign a 25% uncertainty to the parameterisation given in Equation 3. This uncertainty propagates through the suppression estimates. For instance, the uncertainty in the  $u_*$ -related  $K$  enhancement estimate is approximately  $0.6 \text{ cm hr}^{-1}$  (i.e.,  $2.2 \text{ cm hr}^{-1} \times 25\%$ ). The suppression analysis uses  $H_s$  data derived

from ERA5 reanalysis, which likely carry an uncertainty of about 30% in the Baltic Sea (Giudici et al., 2023). Consequently, the uncertainty in the  $H_s$ -related suppression estimate is  $\sim 1.6 \text{ cm hr}^{-1}$  (i.e.,  $\sqrt{(4.0 \times 25\%)^2 + (4.0 \times 30\%)^2} \text{ cm hr}^{-1}$ ). The uncertainty associated with the surfactant-related suppression is substantially larger because it is not directly determined but inferred as a residual after accounting for other components. Combining the propagated uncertainties from the parameterised total  $K$  and from two fetch-induced suppression estimates yields an uncertainty of  $5.8 \text{ cm hr}^{-1}$  (i.e.,  $\sqrt{(22.1 \times 25\%)^2 + 0.6^2 + 1.6^2} \text{ cm hr}^{-1}$ ), corresponding to approximately 110% of the estimated suppression value (Table 1).

Line 377: “ERA” to “ERA5”

Revised.

The suppression analysis uses  $H_s$  data derived from ERA5 reanalysis

Line 383: 4.5 cm/hr here, 4.6 cm/hr in the table. Surely just a rounding error, but worth making them consistent.

Yes, fully agree. Thank you. Revised.

Combining the propagated uncertainties from the parameterised total  $K$  and from two fetch-induced suppression estimates yields an uncertainty of  $4.6 \text{ cm hr}^{-1}$

Line 395: Abbreviation “Chl-*a*” is defined twice in this paragraph. Also, the abbreviation is not used consistently after defining.

Good point. Here we removed the second definition and used Chl-*a* consistently afterwards.

The CenBASE cruise took place during the summer bloom (July), when chlorophyll-*a* (Chl-*a*) is high (Pitarch et al., 2016) and  $f\text{CO}_{2w}$  is strongly reduced by primary productivity (Bittig et al., 2024). To upscale these results, we examine how fetch and surfactants shape the climatological  $\text{CO}_2$  flux of the Baltic Sea. The  $f\text{CO}_{2w}$  indicates a  $\text{CO}_2$  sink in summer and a source in winter (Fig. 7A). However, weaker summer winds and stronger winter winds suggest that the magnitudes of uptake and outgassing may be similar. Seasonal cycles of  $u_*$  and  $H_s$  closely follow wind speed (Fig. 7B), while Chl-*a* peaks during the spring-summer bloom and remains low in winter (Fig. 7C). We estimate monthly surfactant concentrations by scaling the July CenBASE value ( $0.54 \text{ mg L}^{-1}$ ) with

monthly Chl-*a* concentrations following the idea of Wurl et al. (2011) and using the formula  $0.54 \times \text{Chl-}a / \text{Chl-}a_{\text{July}} \text{ mg L}^{-1}$ . Equation 5 is then used to compute the corresponding suppression of gas transfer,  $1 - \frac{1-0.38SA}{0.79} (1 - 0.38e^{-1.25u_*})$ . The resulting *sf* reflects the seasonal Chl-*a* cycle and modulations by  $u_*$ , yielding ~25% suppression in summer and ~10% in winter (Fig. 7C). However, surfactant concentrations are not solely determined by Chl-*a*; for example, humic acids also act as surfactants (e.g., Klavins & Purmalis, 2010), and the Baltic Sea is known for elevated humic acid levels due to significant terrestrial inputs (Hammer et al., 2017). Therefore, estimating surfactants based solely on Chl-*a* has inherent limitations.

Line 418: “especially stronger” to “especially strong”

Revised.

This suppression is especially strong in summer when SA concentrations are highest...

Line 422: “compare” to “compared”

Revised. Thank you

When compared with the open ocean DT-based  $U_{10N}$  formulation...

Reviewer: Brian Butterworth

## Reply to reviewer #2

We sincerely thank Dr. Brent Else for his positive feedback and thoughtful, constructive comments, which have greatly helped us improve the quality and clarity of the manuscript. All coauthors have carefully considered the reviewer's suggestions and worked together to revise the manuscript accordingly. In the responses below, the reviewer's comments are shown in **black**, our replies in **blue**, and the corresponding revisions in the manuscript in **orange**.

I am pleased to present this review of "Mechanisms of air-sea CO<sub>2</sub> exchange in the central Baltic Sea" by Yuanxu Dong and co-authors. The paper presents a well-designed and well-executed field study of gas exchange in an interesting coastal region. Very few similar studies have been conducted, and most of those have occurred (for good reasons) in the open ocean. By focusing this study in the Baltic Sea, important new insights and clarifications about how we should be determining gas exchange in coastal waters are revealed. I thought the manuscript was technically solid. The field methods were well described, and utilized the current state-of-the-art. The choices of data analysis techniques were good, and reasonings used to interpret key factors like the impact of fetch-restricted wave fields and surfactant effects were well justified and acknowledged relevant uncertainties. I really don't have much negative to say about this paper... I think it will be an important contribution to the field, and I support its publication.

Thank you very much for the encouraging feedback and for recognizing the value of our study. We hope that our work will help draw attention within the community to the importance of conducting more gas exchange experiments in coastal environments.

I have a few minor comments that I think the authors should consider addressing before publication:

Line 95: Wave parameters extracted from the ERA5 reanalysis product are an important component of this paper. I think the authors should explain a bit more the uncertainties in these products, and how that may affect their analyses.

This is a good point. In the later stage of the original text, we have included the uncertainty analysis of ERA5 and how it impacts our quantification of the suppression fraction in the

“Uncertainty analysis” section. Below is the relevant text, and hope this solves the reviewer’s concern.

The suppression analysis uses  $H_s$  data derived from ERA5 reanalysis, which likely carry an uncertainty of about 30% in the Baltic Sea (Giudici et al., 2023). Consequently, the uncertainty in the  $H_s$ -related suppression estimate is  $\sim 1.6 \text{ cm hr}^{-1}$  (i.e.,  $\sqrt{(4.0 \times 25\%)^2 + (4.0 \times 30\%)^2} \text{ cm hr}^{-1}$ ).

Line 111: The authors choose to use a 10-minute flux averaging interval, but do not discuss the potential ramifications of this. I would assume it would result in some loss of low-frequency flux. Can this choice be better justified?

Thank you for raising this important point.

For flux calculations, we generally used 20-30 min intervals for the open ocean and 10 min for coastal waters, as coastal environments are more dynamically variable due to their complexity. A shorter time interval may better fit the stationarity requirement, but without losing a significant low-frequency signal. A classical coastal EC study by Vickers & Mahrt (1997) suggests that the wind stress calculated with 5 min averaged over all stationary records captures 95% of the stress values calculated using a 60 min averaging timescale. The stress is not sensitive to the local averaging timescale until the interval is less than 3 min. The 10 min averaging interval we used is also consistent with a previous coastal study (Yang et al., 2016)

Below is a normalized Ogive curve for CO<sub>2</sub> flux from CenBASE, calculated with a 20-minute averaging interval. Low-frequency contributions are negligible, with the majority of flux variance explained by frequencies above  $10^{-2} \text{ Hz}$  ( $\sim 2$ -minute timescale). This characteristic was confirmed across all analyzed time periods.

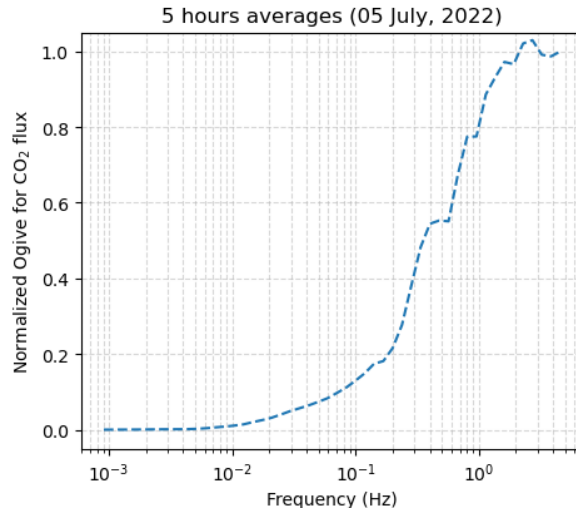


Figure 1 Normalized Ogive curve for EC air-sea CO<sub>2</sub> flux. The results shown here represent an 5-hour average of the 20-min fluxes on 5 July, 2022 during CenBASE.

Vickers, D., & Mahrt, L. (1997). Quality Control and Flux Sampling Problems for Tower and Aircraft Data. *J. Atmos. Oceanic Technol.*, 14, 512–526, [https://doi.org/10.1175/1520-0426\(1997\)014<0512:QCAFSP>2.0.CO;2](https://doi.org/10.1175/1520-0426(1997)014<0512:QCAFSP>2.0.CO;2).

Yang, M., Bell, T. G., Hopkins, F. E., Kitidis, V., Cazenave, P. W., Nightingale, P. D., Yelland, M. J., Pascal, R. W., Prytherch, J., Brooks, I. M., & Smyth, T. J. (2016). Air-sea fluxes of CO<sub>2</sub> and CH<sub>4</sub> from the penlee point atmospheric observatory on the south-west coast of the UK. *Atmospheric Chemistry and Physics*, 16(9), 5745–5761. <https://doi.org/10.5194/acp-16-5745-2016>

Line 190: is this  $u^*$  value actually observed? Just previously, and after this,  $u^*$  from COARE models are discussed. Can you clarify in the text whether this  $u^*$  is coming from eddy covariance, or the COARE model?

Sorry for the confusion. Yes, here is the  $u_*$  from EC observation. We clarify this by rephrasing this sentence as:

Observed values of  $u_*$  from EC momentum fluxes during CenBASE were 10% higher than modelled open ocean  $u_*$  at the same wind speeds (Fig. 3A), likely reflecting fetch-related differences.

Overall, I found it a bit strange that the dual-tracer results were included in this paper. They are not discussed at great length, and apparently there is another manuscript in review (or perhaps

being drafted) that will deal with this in greater detail. I think it is worth keeping the measurements in, but maybe the authors could find a place to more explicitly explain why it is important to have this data in this study.

This is a good point. Yes, the dual-tracer dataset alone has been analyzed in a separate, submitted manuscript, with a preprint forthcoming. There is ongoing debate regarding the suitability of EC for quantifying air-sea gas exchange. Simultaneous application of EC and the dual-tracer (DT) method enables a direct intercomparison of these two approaches. Our results demonstrate that the two methods yield comparable gas transfer velocities under low-moderate wind speeds. However, the EC technique operates on a shorter timescale and provides a wider wind range constraints on the gas transfer velocity. To emphasize this advantage, we have added the following statement to the first paragraph of Section 3.3 (“Gas transfer velocities from EC and DT”):

Simultaneous gas transfer velocity observations by the EC technique and the dual-tracer method provide a direct comparison of the results from these two different approaches.

In the discussion section, the original manuscript also includes a statement:

The  $K_{660}$  derived from both techniques agrees well, confirming the reliability of both methods for gas transfer velocity observations.

Line 218: Appendix A2 is about surfactants, not dual-tracers.

Good spot. Thank you. Revised.

$K_{660}$  from the DT experiment is summarized in Appendix A1.

Line 224 and Line 236: qualitative statements about agreement between DT and EC fluxes are made... I find this a bit weak; there are clearly some DT points that fall below or above the EC fluxes. Can these statements be made a bit more quantitatively?

Good suggestion. From Fig. 4, looks like the several DT points at low wind speed are slightly lower than the EC results, but well within the uncertainty range. For the point at  $U_{10} \sim 7 \text{ m s}^{-1}$ , DT is higher than the EC and the one at  $U_{10} \sim 9 \text{ m s}^{-1}$  is lower than the EC, but considering the uncertainty, they still agree. In general, the DT data is concentrated in the wind speed range of 5-

9 m s<sup>-1</sup> and on average gives 10.3 cm hr<sup>-1</sup> and the CenBASE EC-based parameterisation for the equivalent DT wind speed gives  $K_{660}$  of 11.1 cm hr<sup>-1</sup>. Therefore, the EC result only slightly (7.8%) differs from the DT observations. We took the reviewer's suggestion and gave a quantitative comparison.

Within this wind range, DT-and EC-derived  $K_{660}$  values are in good agreement, with the former being only slightly (~8%) lower than the latter (Fig. 4).

Line 236 is about the comparison of the observed DT results in the Baltic Sea and the modelled DT results based on the open ocean data. Similarly, we added a quantitative comparison.

DT-derived  $K_{660}$  values during CenBASE also generally agree with the open-ocean DT-based parameterisation of Ho et al. (2006) under equivalent wind speeds (orange dashed line in Fig. 4A), with the former on average being only ~7% lower than the latter.

Line 273: I was confused by the statement "applying equation 3 to the equivalent wind speeds"... Equation 3 only includes  $u^*$  and  $H_s$  as variables.

Sorry for the confusion. As also noted by the first reviewer, here we rephrased the sentences:

The observed EC  $K_{660}$  during CenBASE was on average 14.9 cm hr<sup>-1</sup>. To compare this value with open-ocean conditions at equivalent wind speeds, we apply the wind-speed observations from the CenBASE cruise (i.e., wind speed values shown in Fig. 4A) to Equation 3 to estimate open-ocean  $K_{660}$ , yielding average values of  $K_{i660} = 15.1$  cm hr<sup>-1</sup>,  $K_{b660} = 7.0$  cm hr<sup>-1</sup>, and  $K_{660} = 22.1$  cm hr<sup>-1</sup>. This means that the observed  $K_{660}$  during CenBASE was 33% (7.2 cm hr<sup>-1</sup>) lower than the open-ocean  $K_{660}$  estimate.

Section 3.6: The origin of some of the data is a bit unclear... in particular the source of the  $f\text{CO}_2$  data is not well explained.

Good point. We now added the source of the  $f\text{CO}_{2w}$  data in the first paragraph of this section:

To upscale these results, we adopt a  $f\text{CO}_{2w}$  product from Bittig et al. (2024) to examine how fetch and surfactants shape the climatological  $\text{CO}_2$  flux of the Baltic Sea. This climatological  $f\text{CO}_{2w}$  product is derived by combining observations and model patterns.

Figure 7: Why not show us the computed flux? A lot of the text (eg Line 430-434) describes patterns in the computed flux, but this is not presented.

Thank you for this question. The atmospheric  $f\text{CO}_2$  ( $f\text{CO}_{2a}$ ) over the Baltic Sea is not provided by the dataset used for seawater  $f\text{CO}_{2w}$  (Bittig et al., 2024). Flux calculation also requires additional inputs such as sea surface temperature, salinity, and sea ice concentration. Our study does not aim to provide a quantitative flux analysis, but rather to examine how the new parameterization could affect the spatiotemporal patterns of air-sea  $\text{CO}_2$  flux. We therefore believe the current analysis and Figure 7 sufficiently support our conclusions. We hope this clarification addresses the reviewer's concern.

Line 490: There is a reference to a "bubble buoy" which I don't think is described anywhere else in the manuscript? What is this? Is it important to keep in?

A bubble (spar) buoy was deployed, but unfortunately the bubble measurements were not working, but the bubble buoy also integrated temperature, salinity and dissolved oxygen sensors (shown in Fig. A7). Here we just want to highlight the bubble-mediated transfer study requires real bubble dynamic measurements and the bubble buoy is useful to do that. Since the data from the bubble buoy was used, we include a short description in the section 2.3 (**Auxiliary observations**) to raise the reader's awareness that a buoy was deployed during the cruise:

A spar buoy equipped with cameras, temperature, salinity, and dissolved oxygen sensors was deployed at several stations to characterize upper-ocean bubble and water column dynamics.