

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 CO<sub>2</sub> 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 an 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:

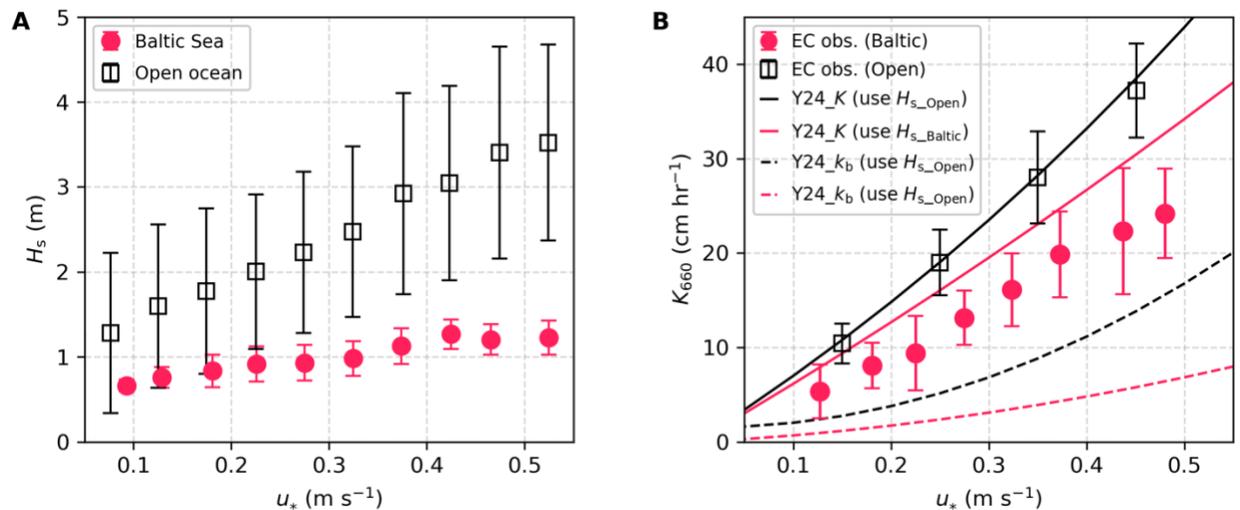


Figure 5: Comparison of significant wave height ( $H_s$ ) and  $K_{660\_CO2}$  between the Baltic Sea and the open ocean. A:  $H_s$  in the Baltic Sea during CenBASE (red dots) and in the open ocean (black squares),

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<sup>-1</sup>)” 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