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Authors: Lucía Gutiérrez-Loza et al.
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Authors' reply to Anonymous Referee #2

Dear reviewer,

We would like to thank you for reviewing this manuscript. We have addressed your comments and provide with the answers below. We consider that the manuscript has improved significantly based on you feedback, and we are grateful to you for that.

The response to each comment is written in blue italics, *while the changes made in the revised manuscript are in red*.

This manuscript by Gutiérrez-Loza et al. presents a new dataset of eddy correlation air-sea CO2 flux measurements that is in itself a valuable contribution to the field. Analysis of the dataset has the potential to improve our understanding of gas exchange processes and the analysis done here does reveal some new insights particularly regarding the importance of water-side controls on the gas exchange rate and the influence of processes that cannot be parameterised as a function of wind speed. It is generally well written, interesting and easy to follow. I encourage the authors to work further on the dataset to see it through to publication.

However, there are a few major issues that would be essential to address first:

Major issues:

- How representative are the results here in a global context? The title leads one to expect that these will be universally applicable insights, but this does not seem to be the case. Indeed the most confidence the authors were able to express in the wider applicability of their results was 'the results presented here are most probably relevant for other marginal seas and coastal areas' (line 375, emphasis mine). Line 314 also suggests this work may be only relevant to the Baltic. The title should be further qualified '… in the Baltic Sea' or similar unless the authors can be sure that their results are more widely applicable.

The approach used in the revised manuscript (see replies further down in this document) proved to be a more suitable, and statistically robust, method for the analysis of k_660. The results from this approach were used to identify conditions enhancing the gas exchange. We further linked such conditions to the potential effect of sea spray (under high wind speeds) and water-side convection (under low wind speed conditions). We consider these two mechanisms to be potentially relevant in other regions and spatio-temporal scales. Detailed analysis of the wider *applicability of these results is beyond the scope of this work. However, the potential relevance of these mechanisms is discussed in the revised manuscript.*

*The **title** of the manuscript was changed to "**On physical mechanisms enhancing air-sea CO2 exchange**".*

*Discussion about the potential relevance of sea spray and water-side convection in other regions was included in the **Discussion** section.*

- One of the key motivations for this study is reducing uncertainties in gas exchange calculations (e.g. lines 22 – 24) yet there is no meaningful uncertainty analysis of the results of this study. I could not even see uncertainty estimates for the raw measurements that underlie the new dataset being presented and there was no propagation of uncertainty through to the final results. This is essential especially if the results are to be compared with previous work or other approaches, else you cannot be sure if the results are actually consistent or not.

In this sentence (lines 22-24) we acknowledged the existence (and relevance) of the uncertainties associated to the air-sea CO_2 fluxes at a global scale. Furthermore, we stated that a large proportion of these uncertainties exist due to the "incomplete understanding of the spatio-temporal variability in the controlling mechanisms". The intention of this sentence is not to set up the exact focus of current study, but rather to put into context why more process-oriented studies are necessary, and the implications in the global context.

The focus of our study is to capture the temporal variability of k_660 and other processes involved in the exchange. We assessed this by presenting long records of high-frequency data, thus, capturing both the short- and long-term (several years) variability of FCO_2 and k_660. In the revised manuscript, we presented a more statistically robust analysis of the data and used it to identify mechanisms that can potentially cause large deviations on k_660.

A subsequent sentence was included in the first paragraph of the introduction. This with the aim of highlighting the relevance of resolving the different mechanisms involved in the gas exchange:

"However, large uncertainties are still associated with the air--sea CO_2 flux estimates, mainly due to the incomplete understanding of the spatio-temporal variability in the controlling mechanisms. **Resolving the effect of these mechanisms at the relevant temporal and spatial scales is essential to constrain the oceanic contribution in the global carbon balance**."

- Many of the relationships described in section 3.2 and its subsections were not convincing based on the figures. For example on line 225 'kr showed a clear relationship with significant wave height … (Fig 6a)' but if we look at Fig 6a, then I see more clearly the colours getting 'higher' in more vertical bands towards the right (ie correlating with U10) rather than vertical bands towards the top. Same applies for line 239 comment about mixed layer depth. But in general I think that the format of Figs 6-10 makes it very hard to see the correlations described anyway: you are trying to eyeball the angle at which changes in colours occur and there are so many datapoints that they all block each other (e.g. fig 8b has a section in the middle that looks all blue i.e. low values, but with hints of higher pink points that can just be seen around the edges – very hard to interpret). My suggestion would be to replot these figures with kr on the x-axis and the variable of interest (currently the colours) on the y-axis. The points could then be coloured by U10 or something else. This would be a much more clear and convincing way to see correlations. Furthermore, related to point (2) above, there needs to be statistics for the correlations that you report.

We thank Reviewer 2 for this comment. Based on this and other comments received on the original submission, we decided to reframe the way the analysis was made and, as suggested here, the way the data is presented in the revised manuscript. Firstly, the data is no longer analyzed using the residual gas transfer velocity (k_r). Instead, we use a normalized gas transfer velocity for each 30-min average, defined as k_660/k_wind, where k_wind is the best fit to the bin-averaged k_660 values of the current study. Secondly, the normalized gas transfer velocity was plotted (for each wind speed regime) as a function of each of the variables of interest (i.e. ΔpCO2, MLD, Hs, etc.). We consider that, given the complex relationship between the wind speed and the other parameters evaluated in this study, it would be hard to find meaningful correlations between k_660 and those parameters. However, based on the analysis of the normalized gas transfer velocity, it was possible to identify a set of conditions leading to deviations of k_660 from k_wind. The analysis was carried out using boxplots which provide statistical summaries of the data. Subsequent analysis was then made based on the behavior of k_660 under the enhanced conditions found.

Figures 6 to 10 were substituted by the following figures:

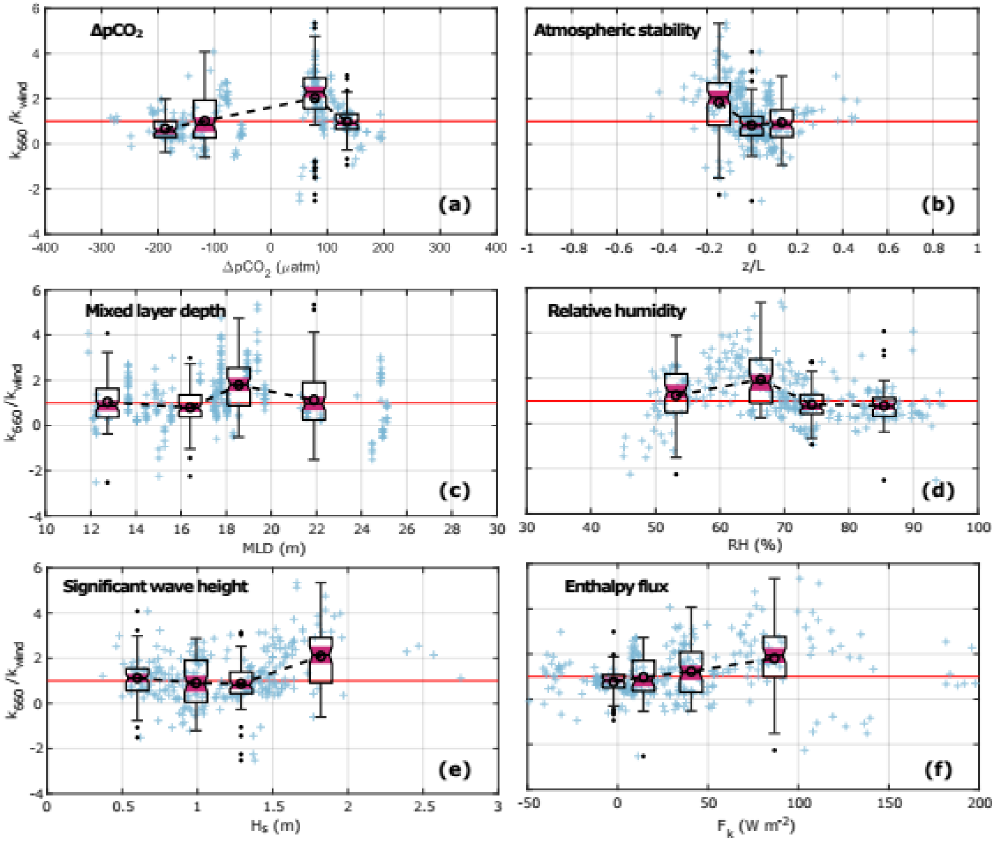

Figure 6. Normalized gas transfer velocity (k_{660}/k_{wind}) under high wind speed conditions ($U_{10N} > 8\,\mathrm{m\,s^{-1}}$) as a function of a) ΔpCO_2, b) atmospheric stability (z/L), c) mixed layer depth (MLD), d) relative humidity (RH), e) significant wave height (H_s), and f) total enthalpy flux (F_k). The crosses represent the individual half-hourly values. The boxplots give a statistical summary for equidensity bins defined based on the distribution of k_{660}/k_{wind} as a function of each of the parameters (see Appendix A). The median, first, and third quartiles are represented in each box; the whiskers represent the minimum and maximum values, and the black dots represent the outliers; the notches highlighted in pink indicate the median's 95 % confidence interval. The open circles linked with a dashed line indicate the bin means, and the horizontal red line indicates $k_{660}/k_{wind} = 1$.

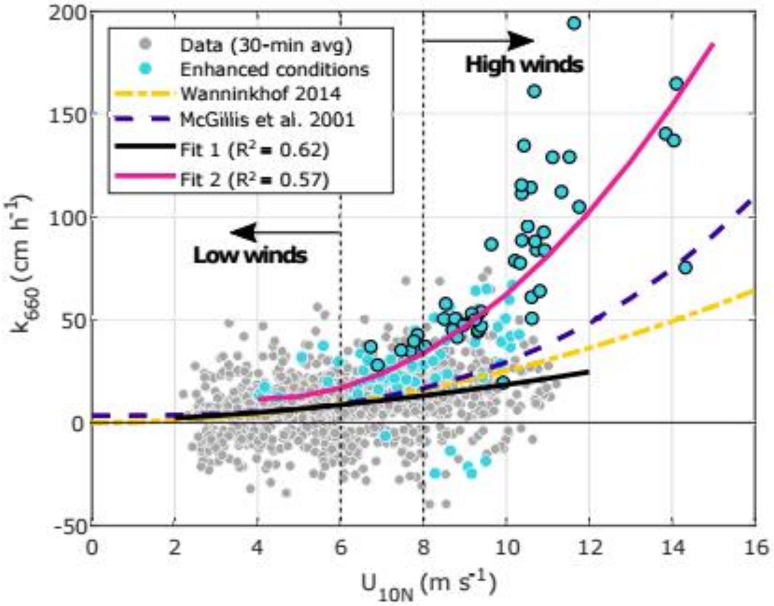

Figure 7. Gas transfer velocity for CO_2 (adjusted to a Schmidt number of 660) as a function of the 10 m neutral wind speed. The dots represent the half-hourly values of k_{660}. The blue dots represent k_{660} under enhanced conditions (see text for details), while the blue dots with a black edge indicate cases where $H_s > 1.5$ m. The black line represents the best fit (quadratic) to the data excluding the enhanced cases (only gray dots), while the pink line is the best fit to the enhanced data (only blue dots). For reference, a quadratic (Wanninkhof, 2014) and cubic (McGillis et al., 2001) wind-based parametrizations were included. The wind speed regimes are separated by vertical dashed lines.

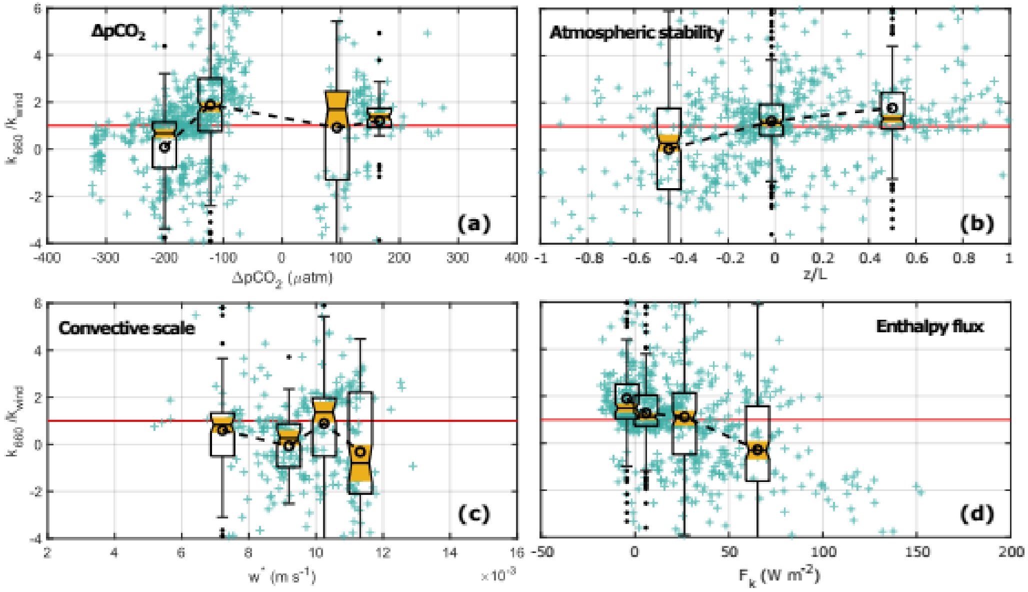

Figure 8. Normalized gas transfer velocity (k_{660}/k_{wind}) under low wind speed conditions ($U_{10N} \leq 6\,\mathrm{m\,s^{-1}}$) as a function of a) ΔpCO_2, b) atmospheric stability (z/L), c) water-side convective scale ($w*$) under unstable atmospheric conditions, and d) enthalpy flux (F_k). The crosses represent the individual half-hourly values. The boxplots give a statistical summary for equidensity bins defined based on the distribution of k_{660}/k_{wind} as a function of each of the parameters (see Appendix A). The median, first, and third quartiles are represented in each box; the whiskers represent the minimum and maximum values, and the black dots represent the outliers; the notches highlighted in yellow indicate the median's 95 % confidence interval. The open circles linked with a dashed line indicate the bin means, and the horizontal red line indicates $k_{660}/k_{wind} = 1$.

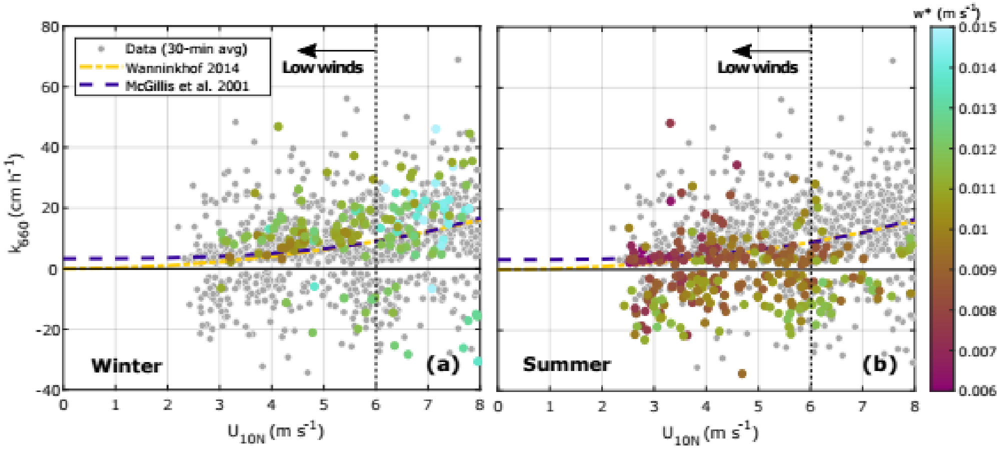

Figure 9. Gas transfer velocity for CO_2 (adjusted to a Schmidt number of 660) as a function of the 10 m neutral wind speed during a) winter and b) summer. The dots represent the half-hourly values of k_{660}. The color represents the water-side convective scale ($w*$) for data under unstable atmospheric conditions, calculated according to Rutgersson and Smedman (2010). The wind speed regimes are separated by a vertical dashed line.

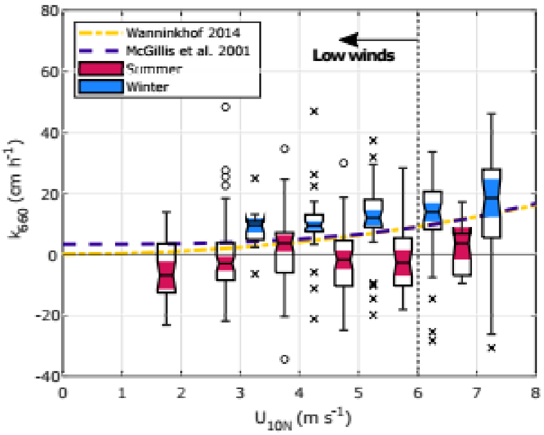

Figure 10. Boxplots of the gas transfer velocity for CO_2 (adjusted to a Schmidt number of 660) during unstable atmospheric conditions as a function of the 10 m neutral wind speed during summer (pink) and winter (blue). The median, first, and third quartiles are represented in each box; the whiskers represent the minimum and maximum values, and the circles and crosses represent the outliers; the notches highlighted in color indicate the median's 95 % confidence interval. The wind speed regimes are separated by a vertical dashed line.

- Many parts of the dataset are excluded and the impact of this on the results and their wider applicability is not much discussed. We have low wind speeds on line 139, low fluxes on 154, high humidity on line 157, stratified conditions on line 169, and unexplained low k660 values on line 174. Maybe it's valid to not include these in the analysis, but we really need an accompanying robust discussion of how often those conditions occur in the real world and what that means for the gas exchange rate.

*We agree that further clarity and transparency was necessary when discussing the quality control and the amount of data rejected, as well as the implications of rejecting such data from the analysis of the gas transfer velocity. This issue was tackled by including a section (**Appendix B**) describing the relative importance of each quality control criterion.*

***Appendix B** was included where a more detailed description of the relative importance of every QC criterion (i.e. the effect of each criterion on the total amount of data) is presented and brief discussion about the final size of the data set.*

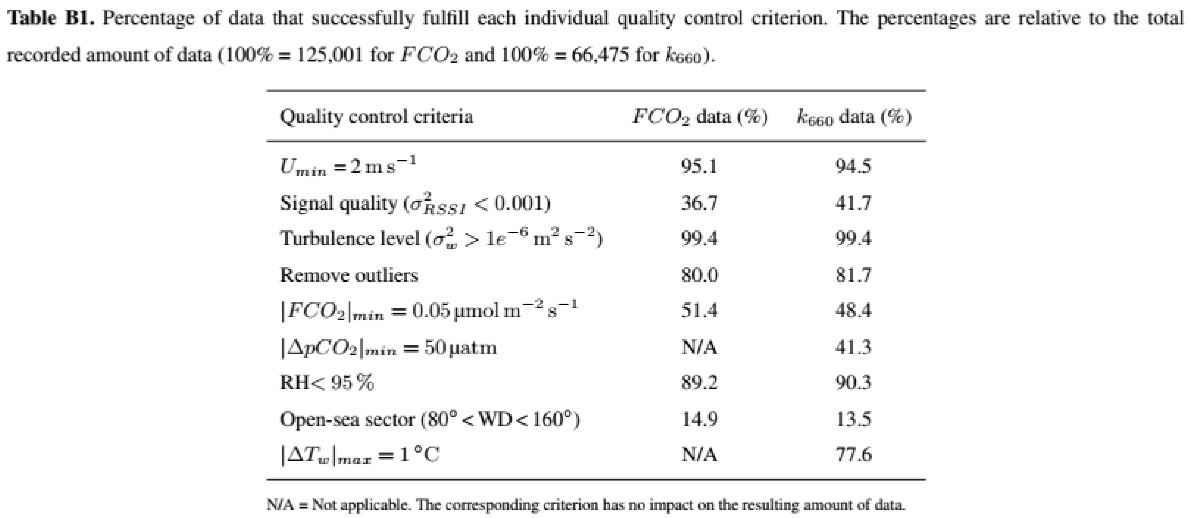

Table B1. Percentage of data that successfully fulfill each individual quality control criterion. The percentages are relative to the total recorded amount of data (100% = 125,001 for FCO_2 and 100% = 66,475 for k_{660}).

Quality control criteria	FCO_2 data (%)	k_{660} data (%)
$U_{min} = 2\,\mathrm{m\,s^{-1}}$	95.1	94.5
Signal quality ($\sigma^2_{RSSI} < 0.001$)	36.7	41.7
Turbulence level ($\sigma^2_w > 1e^{-6}\,\mathrm{m^2\,s^{-2}}$)	99.4	99.4
Remove outliers	80.0	81.7
$\lvert FCO_2\rvert_{min} = 0.05\,\mu\mathrm{mol\,m^{-2}\,s^{-1}}$	51.4	48.4
$\lvert \Delta pCO_2\rvert_{min} = 50\,\mu\mathrm{atm}$	N/A	41.3
RH< 95 %	89.2	90.3
Open-sea sector (80° < WD < 160°)	14.9	13.5
$\lvert \Delta T_w\rvert_{max} = 1\,°\mathrm{C}$	N/A	77.6

N/A = Not applicable. The corresponding criterion has no impact on the resulting amount of data.

Furthermore, an initial evaluation of the probability distributions of several parameters included in the analysis showed that the initial data set (before quality control) and the final data set (after quality control) had similar patterns. Thus, indicating that the QC-ed data set used in the k_660 analysis was representative of the local conditions and biases due to the exclusion of data are expected to be small, if any. This analysis was not included in the manuscript but figures can be found in the response to the comments of Bernd Jähne (community comment 1).

Brief discussion of the implication of removing data under certain criteria (e.g. U10N<2 m/s), was included as part of the ***Discussion*** *section.*

- Despite high variability and physical/biogeochemical heterogeneity being an important motivator of the study, some key properties were assumed to be uniform (salinity on line 116, somewhat cryptic ‘biogeochemical water properties’ on line 101). This may be fine but this assumption is not critically assessed. One should be able to quantify a maximum effect size for how important ignoring this variability could be.

We acknowledge the fact that there is a significant variability in the biogeochemical properties, however, some assumptions had to be made during the analysis due to practical reasons. We believe, however, that this assumptions are well founded. For instance, the use of a constant salinity value for the solubility calculations. As stated in the manuscript, salinity values range between 6.5 and 7.5 PSU in the area of study, which in turn have a small impact on the CO_2 solubility when compared with the effects of temperature (see Weiss, 1974), which also presents a much larger variability in the current site. Furthermore, the final effect of salinity on the gas transfer velocity calculations is of the order of $1\text{x}10^{-2}$ *cm/h per PSU (based on an initial sensitivity analysis). Such effect was considered to be negligible given the low salinity range.*

References to ***Wesslander et al., 2010*** *and* ***Rutgersson et al., 2020*** *were included. In that study, long records of sea surface salinity in the central Baltic Sea show its limited variability.*

On the other hand, the assumption about the homogeneous biogeochemical water properties is based on the wind-direction classification discussed in Rutgersson et al, 2020. This *consideration was made in order to be able to assume that the seawater pCO_2 measurements (recorded at a single location) were representative of the entire open-sea sector; furthermore, that the fluxes measured at the tower can be associated to these seawater measurements. In* ***section 3.2.2*** *we stated that "the strong stratification, relatively weak ΔpCO_2, and the possibility of strong heterogeneity in terms of the biogeochemical properties might hinder our capacity to calculate k_660 from pCO_2w and FCO_2". We, therefore, suggest that "the interpretation of these data should be taken with some caution".*

A detailed assessment about the wind-direction categories, including the analysis of the water properties in the region is presented in Rutgersson et al., 2020. For such analysis, observations of temperature, salinity, pCO2, dissolved oxygen, pH, chlorophyll-a, and nutrients in the vicinity of Östergarnsholm were taken into account.

In ***Section 2.1.1****:*

"Furthermore, the biogeochemical water properties **and the hydrographical features** were assumed to be **spatially** homogeneous **along** this sector (Rutgersson et al., 2008, **2020**), **ensuring that the water-side measurements were representative of the flux footprint of the tower.**"

Other minor comments:

46 how deep is 'the upper layer of the ocean'

The upper layer of the ocean can be considered to be the layer that is adjacent to the air-sea interface and that is affected by processes at the ocean surface (i.e. meteorological conditions, wave field, radiation, etc.). However, there is no universal definition of what the depth of this layer is, being strongly dependent on the local and regional conditions. In general, the depth of the upper layer of the ocean could be considered to be from a few meters to several tens of meters (e.g. Soloviev and Lukas, 2013, Moum and Smyth, 2019).

The sentence was modified to:

"At moderately high wind speeds, above 8-10 m s-1, the upper layer of the ocean is generally well mixed **(from the surface up to several tens of meters depth)**."

Fig 1 caption 'see text for details' of open sea sector – please give section number and repeat values here

Changes were made accordingly.

Caption in ***Figure 1*** *was modified to:*

"(a) Map of the Baltic Sea; the red cross in the central Baltic Sea indicates the location of the Östergarnsholm station. (b) Map of the Östergarnsholm station ca 4 km off from the Gotland Island; the red dot indicates the location of the tower, the blue cross is the location of the mooring with water-side instrumentation (Sect. 2.1.2), and the shaded blue area is the so-called "open-sea" sector **with wind directions from 80°< WD < 160°** (see **Sect. 2.1.1** for details)."

Fig 2 Very unclear to me what this figure shows. What are X and Y on the axes? What does ‘footprint’ mean?

The flux footprint, f(x,y), is a transfer function used to relate the sources or sinks of scalars at the surface with the measurements made at a specific height (Kljun et al. 2015). The footprint represents the contribution per unit area of each unit (i.e. m^2) source or sink ($Q_u(x,y)$) to the total flux measured ($F_c(0,0,z)$), and the units are expressed in m^{-2}:

$$f(x,y) = \frac{F_c(0,0,z)}{Q_u(x,y)} = \left[\frac{\mu mol\ m^{-2} s^{-1}}{\mu mol\ s^{-1}}\right] = [m^{-2}]$$

In Figure 2, the average footprint of the fluxes at the tower is presented for different atmospheric stability conditions. There, X and Y are length scales in meters using the reference point (0,0) as the tower where the measurements are made.

Some clarifications were made in the manuscript, however, an in-depth description of this concept is beyond the scope of this work.

A brief description of the concept of footprint was included in ***Section 2.1.1 (Atmospheric data)****:*

“The flux footprint is a function used to characterize the contributions of the sources and sinks per unit area to the total flux measured at a certain point. Based on this mathematical concept, it is possible to associate the fluxes measured at a specific height with the surface exchange of any scalar (Kormann and Meixner, 2001)”

A more concrete description of the content of ***Figure 2*** *is now given****:***

“Figure 2 shows the spatial distribution of the flux contributions (in m^{-2}) for different atmospheric stability conditions”

Additional information about the color scale in the figure is given in the caption of ***Figure 2:***

“Average footprint distribution for (a) unstable, (b) neutral, and (c) stable atmospheric conditions. The green cross indicates the position of the tower and the contours represent the percentage of source area from 10-80%. **The flux footprint (in color) shows the spatial distribution of the contributions per unit area to the total FCO2 (in m^{-2})**. The footprint was calculated using the model developed by Kljun et al. (2015) using all data available for the open-sea sector between mid-2013 and 2020”

161 ‘in these region’ => ‘in this region’

The error was corrected.

175 ‘more detail analysis’ => ‘more detailed analysis’

The sentence has been removed from the text. In the revised manuscript, the negative k values were included in the analysis, thus, the sentence was no longer needed.

183 mixed up > and < symbols for intermediate conditions

Thanks for pointing this out. The error was corrected.

Section 3.1 worth pointing out that the seasonal cycle of pCO2w looks to be biologically controlled rather than temperature controlled in this part of the world – any impact on wider applicability? See e.g. analysis of Takahashi et al (2009, DSR2)

In addition to the analysis presented by Takahashi et al., 2009, there have been studies suggesting that the seasonal cycle of pCO2w is, to a large extent, controlled by biological activity in the Baltic Sea (e.g. Helmuth and Schneider, 1999, Wesslander et al., 2010). The seasonality occurs in both, the local biological activity and the transport of organic matter. A detailed analysis of these processes is beyond the scope of this study. However, we agree with Reviewer 2 and we, therefore, mention the relevance of the biological activity on the seasonal cycle of pCO_2w.

*The following sentence was included in **Section 3.1:***

*“*The seasonality in pCO_2w in the Baltic Sea has been recognized to be strongly modulated by the biological activity (Helmuth and Schneider, 1999, Wesslander et al., 2010)”

Fig 3 State which method of calculating air-sea CO2 fluxes is used here

Text was modified accordingly.

*Caption in **Figure 3** was modified to:*

“Annual cycle of (a) CO2 partial pressure (pCO2) in the seawater and in the atmosphere, and (b) air–sea CO2 fluxes **from eddy covariance**. The dots represent the half-hourly values while the solid lines show the monthly averages.”

228 please explain briefly why the wave age suggests local generation of waves (for the non-expert)

The wave age is a measure of the effect of the wind over the wave field. Therefore, when the ratio between Cp and U10N is small, there is an indication that the effect of the wind forcing on a specific wave group is large, thus, generating wave growth. On the contrary, when this ratio is large, the wave field is developed and the forcing of the wind has little effect on the waves.

A brief explanation about why the observed wave age values suggests locally-generated waves was included:

“While **the small values of** the wave age (C_p/U_{10N}, Fig. 6c) **suggest wave growth caused by the forcing of the wind over wave field. Thus, indicating** locally generated waves (i.e. wind sea) at these wind speed conditions.”

Figs 4 & 5 suggest to make points smaller and semitransparent so that structure within the big grey overlapping blob can be seen

Thank you for the comment. We agree that the data can be tightly clustered and overlapping in some areas, making it difficult to visualize individual data points. Changes were made in Figure 5 to avoid this issue and improve the visualization of the data as much as possible. However, for Figure 4, the amount of data is significantly higher (less data are removed during quality control for these variables in comparison with k660). Visualization of individual data points is hard with such amounts of data, thus, we considered that showing the figure as is still give a good-enough understanding of the general behavior of the data.

*Changes in **Figure 5** were made. The dots were made smaller and with white edges to make it easier to see when dots are overlapping. Additional color shading was included to indicate the data density. The legend was modified accordingly:*

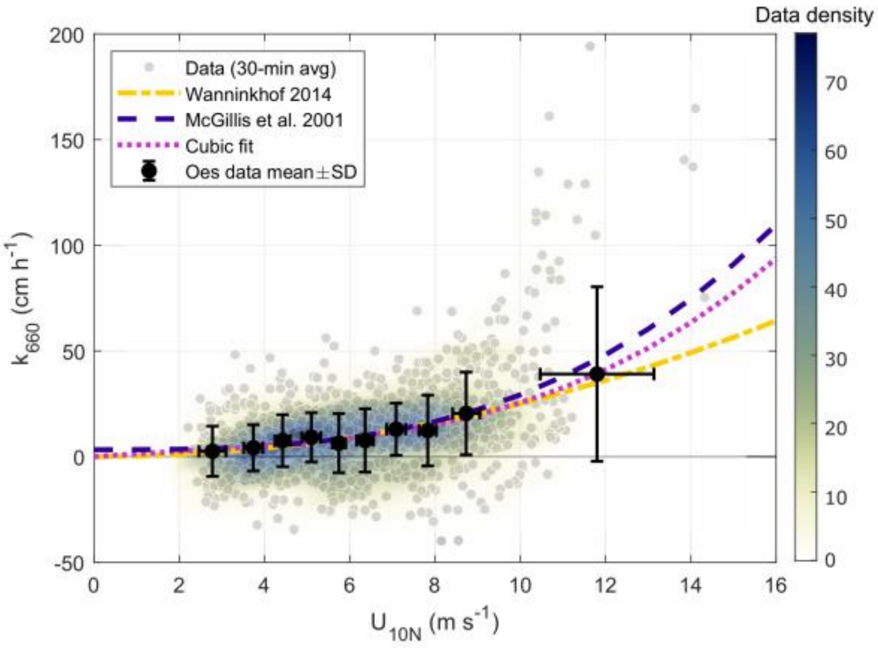

Figure 5. Gas transfer velocity for CO_2 (normalized to a Schmidt number of 660) as a function of the 10 m neutral wind speed. The grey dots represent the half-hourly values of k_{660} for the nine-year period from 2013 to 2021. The black dots and bars, represent the k_{660} mean values and standard deviations, respectively, calculated for equi-density bins based on the wind speed percentiles; the best fit to the means is shown as the pink dotted line. For reference, a quadratic (Wanninkhof, 2014) and cubic (McGillis et al., 2001) wind-based parametrizations were included.The colors in the shaded area represent the data density (in counts) with a grid bin size of 1 m s^{-1} by 10 cm h^{-1}.

300 there are studies that compare these other parameterisations, have you looked at those to put your comparison in more context?

The quadratic (W14) and a cubic (Mc01) parametrizations were included in the manuscript as a reference. However, further analysis of these, or other, wind-based parametrizations was beyond the scope of this study.

305-306 not convinced that this long-term average being correct but short term was really ‘shown’ here. Needs statistics and more rigorous definitions (what is long term? How much uncertainty is there by ignoring the water side effects?)

We have decided to keep the wind-based as a (visual) reference, thus, no further comparison was made between our data and these parameterization. With this in mind, we decided to remove the statements in which we suggested that existing wind-based parameterization might be good representations of the gas exchange in the study region. Instead, we developed the discussion around the k_wind, which was obtained from data presented in this study. Furthermore, we avoided the use of long-term (in the context referred here) and instead use the words "average" or "bin-averaged".

We would like to point out that we are not "ignoring the water side effects" in the analysis by using EC data. On the contrary, by using EC we are directly measuring the total flux, thus, accounting for all the processes involved in the transport (even if these are not measured individually). We apologize if we are misunderstanding this question.

394 define ‘adequately’ (adequate for what purpose?)

This particular phrase was removed. We agree that further comparison between our data and existing wind-based parameterizations would be necessary in order to find out how adequate these parametrizations are for this study site.

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