MS No.: bg-2022-82
Authors: Lucía Gutiérrez-Loza et al.
https://doi.org/10.5194/bg-2022-82

Authors' reply to Referee #1 Bryce Van Dam

Dear Bryce,

We would like to thank you for taking the time of reviewing this manuscript and for the very thorough and useful comments provided. We have made our best to address each of them, and we consider the manuscript to have improved significantly based on your feedback. Please find below the responses to each of your comments.

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

- **Primary concerns:**

First, I am not certain that residual k (k_r) formulated in this manner (k_measured – k_W14) indeed has the effect of "removing the wind-speed dependency from k_660" (line 177, interpretations thereafter). This is because the W14 parameterization was developed for the ocean basin scale, using a (revised) estimate of the bomb 14C inventory and a global wind product. As described in the comments and recommendations section of W14, this formulation is intended to be used for "regional-to-global flux estimates of CO2". The W14 formulation is also intended for longer (multiple hours) time scales by squaring wind speed and averaging over 6+ hours, so I am not sure this is appropriate to compare with the 30-minute averaging intervals used in GL. Put simply, W14 describes the global relationship between wind and k, but does not necessarily isolate the effect of wind speed on k (except for the y-intercept which is forced through 0). Therefore, while k_r as formulated should correct for some of the wind-speed dependency from k_660, I do not feel that it can do so comprehensively at the time-scale applied here.

With this being said, I am open to the use of k_r in this way, provided that the authors:

1) Disagree with my explanation above and can provide a reasonable rebuttal explaining so, or
2) If they decide to revise the manuscript text to verbally describe this issue, or
3) If they can calculate a new k_r in a way that better incorporates the uncertainties in the wind-based parameterization (W14 here).

We understand the concerns regarding the suitability of W14 to remove the "wind-speed dependency" from our data and we agree with the explanation given. Therefore, we have reconsidered the use of a residual gas transfer velocity (k_r) for the analysis. In the revised manuscript, the W14 parametrization is only shown as a reference but not included as part of the *calculations. Instead, we used a normalized gas transfer velocity defined as k_660/k_wind, where k_wind was obtained from the cubic relationship fitted to the equidensity bin averages of the data set of the current study (shown in pink dashed line **Figure 5**). This normalized gas transfer velocity was used, then, to identify conditions with gas transfer velocities higher-than-expected solely by the k_660 vs U10N relationship under the different wind speed regimes. Subsequent analysis was carried out on k_660 itself.*

*Updated version of **Figure 5**:*

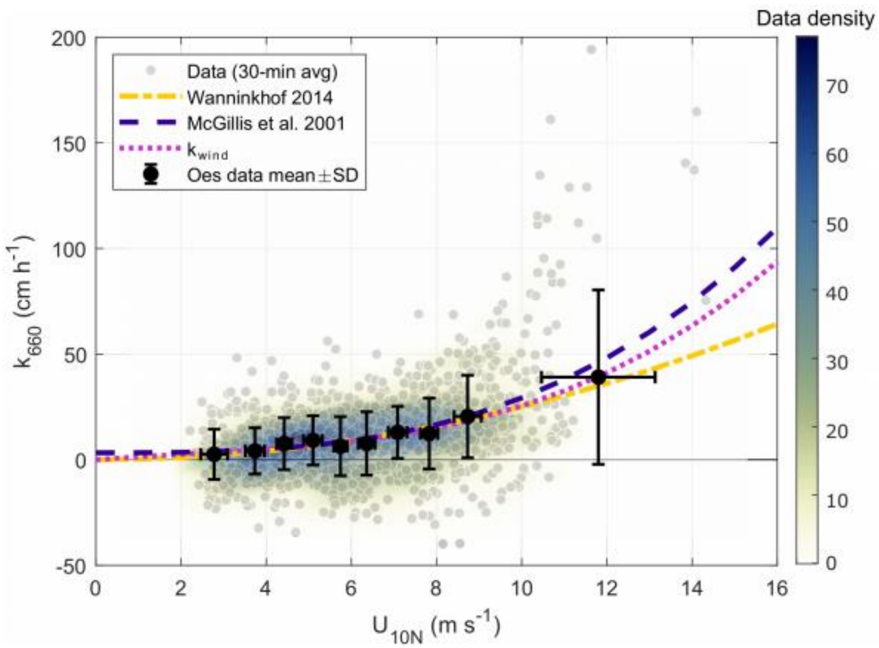

Figure 5. Gas transfer velocity for CO_2 (adjusted to a Schmidt number of 660) as a function of the 10 m neutral wind speed. The grey dots represent the half-hourly values of k_{660} for the nine-year period from 2013 to 2021. The black dots and bars, represent the k_{660} mean values and standard deviations, 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 (k_{wind}). 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}.

***Data processing section (2.2)**. Information regarding k_r was removed and the following paragraph was included:*

"The calculated k_660 were used to study the effect of water-side and atmospheric control mechanisms on air-sea CO_2 exchange. A wind-speed relationship (k_wind) was calculated as the cubic (best) fit to the bin-averaged k_660, using equidensity bins of the wind speed, and used to obtain a normalized gas transfer velocity defined as k_660/k_wind."

*Corresponding changes in the **Results** section, from **sub-section 3.2** and onwards, as well as in the **Discussion.** Particularly, Figures 6 to 10 were removed and substituted by the following figures, and corresponding description and discussion (not presented in full here). **Figure 6** shows the normalized gas transfer velocity vs. several parameters for the high wind speed regime, where enhanced values of k_660 can be identified (the full set of figures was included as*

***Appendix A** in the revised manuscript). Based on this, we selected a data set with particular conditions: positive* $\Delta pCO2$*, strong mixing, high significant wave height, dry air and unstable atmospheric stratification. This feature set was found to be associated with higher k_660 than predicted by the wind speed relationships **(Figure 7)** in what we suggest can be the effect of sea spray**.** A brief excerpt from the text presenting these results, reads as follows:*

"Based on the analysis presented in Fig. 6, we identified a set of conditions that seemed to be associated with enhanced values of k_660. These conditions were characterized by positive ΔpCO_2, strong water-side mixing and dry air (RH < 70 %) during unstable atmospheric stratification. A wave field with H_s >1.5 m further enhanced the gas exchange. Gas transfer velocities higher than predicted, not only by k_wind, but also by other commonly-used parametrizations were observed under these specific conditions (Fig. 7). These enhanced conditions, were observed particularly during high wind speeds, but also during the intermediate regime, and to a much lesser extent during the low wind speed conditions. When these data was removed from the analysis, k_660 seemed to be better represented by U_10N following a quadratic relationship (R^2 = 0.62). The enhanced k_660 (blue dots in Fig. 7), showed a wind-speed dependency of higher order (cubic) and R^2 = 0.57."

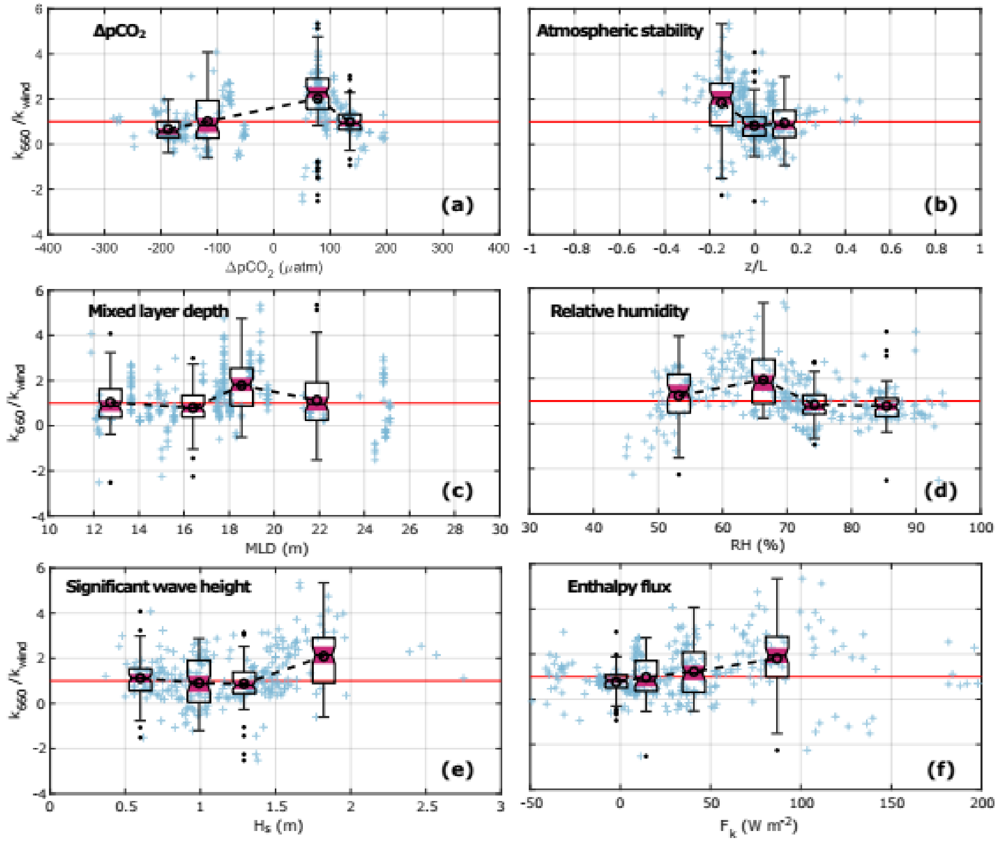

Figure 6. Normalized gas transfer velocity (k_{660}/k_{wind}) under high wind speed conditions ($U_{10N} > 8\,\mathrm{m\,s^{-1}}$) as a function of a) ΔpCO_2, b) atmospheric stability (z/L), c) mixed layer depth (MLD), d) relative humidity (RH), e) significant wave height (H_s), and f) total enthalpy 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 $k_{660}/k_{wind} = 1$.

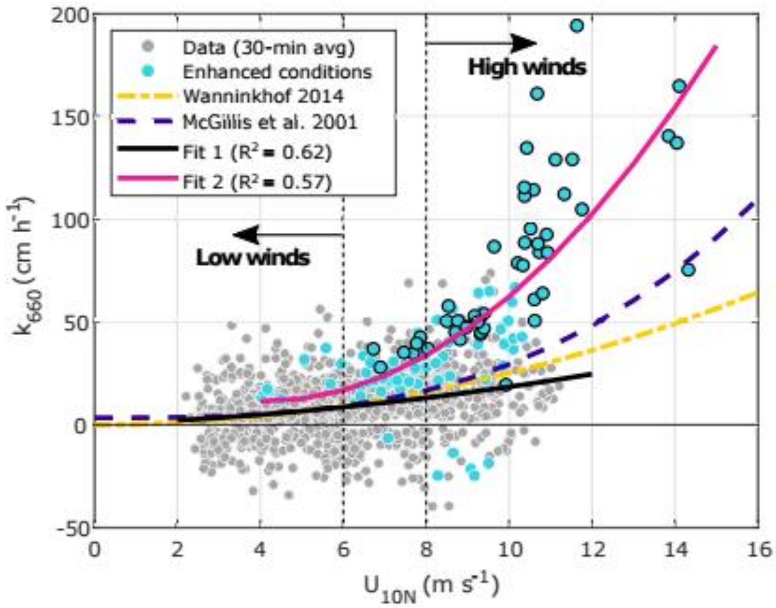

Figure 7. Gas transfer velocity for CO_2 (adjusted to a Schmidt number of 660) as a function of the 10 m neutral wind speed. The dots represent the half-hourly values of k_{660}. The blue dots represent k_{660} under enhanced conditions (see text for details), while the blue dots with a black edge indicate 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.

*Similarly, **Figure 8** shows the normalized gas transfer velocity (k_660/k_wind) for the low wind speed regime. Large variability was observed under unstable conditions and large water-side convective scales. Further analysis of the data under these conditions was explored and presented in **Figure 9** and **Figure 10**, where higher k_660 were observed during the winter times, with higher w* values at low and intermediate wind speeds compared to the summer. A brief excerpt from the text presenting these results reads as follows:*

"Further analysis of the effect of water-side convection on k_660 showed that this process can enhance the gas exchange under unstable atmospheric stratification. Particularly during winter, when persistent cooling of the sea surface was expected (ΔT>0), enhanced values of k_660 were observed at low and intermediate wind speeds associated to high values of w* (Fig. 9a and 10). On the contrary, low values of w* were predominant during the summer months, and linked to low values of k_660 (Fig. 9b and 10)."

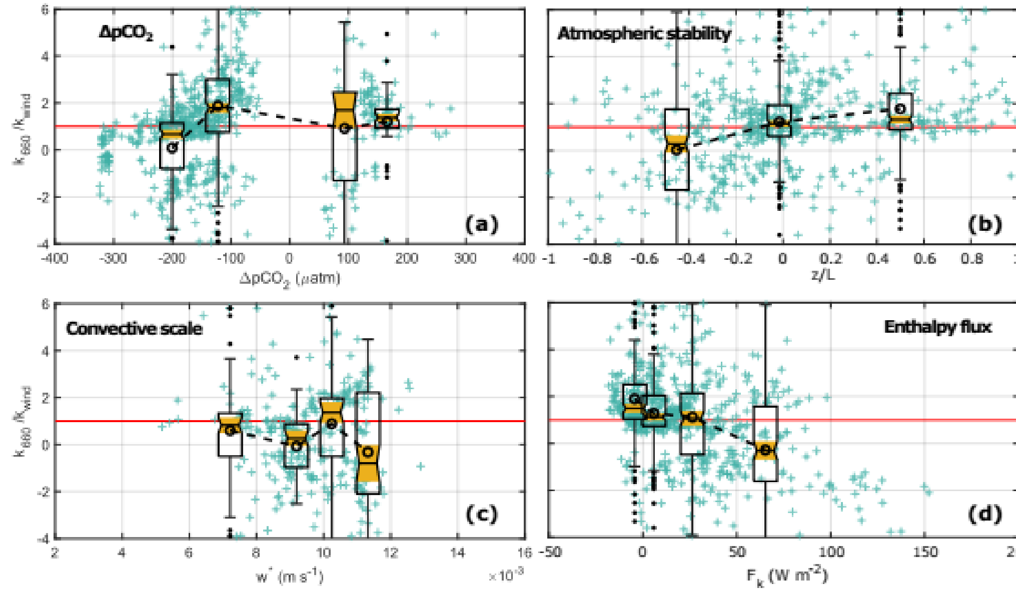

Figure 8. Normalized gas transfer velocity (k_{660}/k_{wind}) under low wind speed conditions ($U_{10N} \leq 6\,\mathrm{m\,s^{-1}}$) as a function of a) ΔpCO_2, b) atmospheric stability (z/L), c) water-side convective scale ($w*$) under unstable atmospheric conditions, and d) enthalpy flux (F_k). The 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 $k_{660}/k_{wind} = 1$.

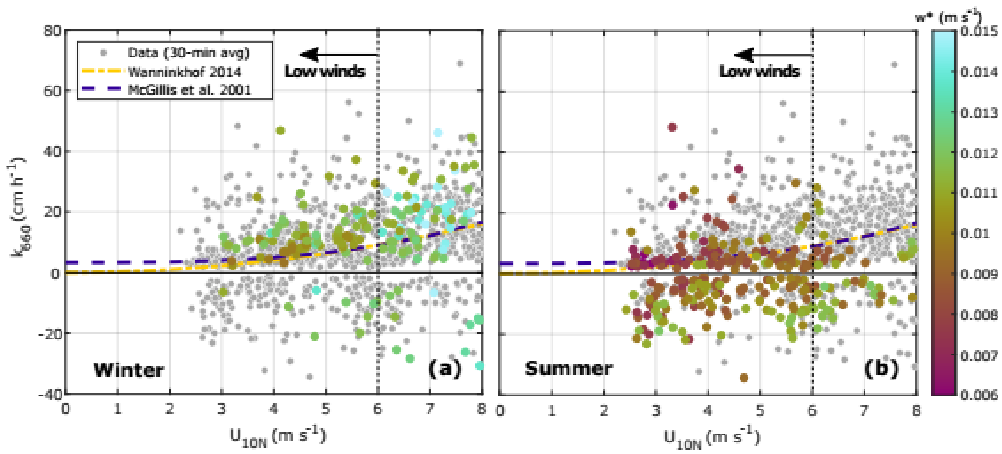

Figure 9. Gas transfer velocity for CO_2 (adjusted to a Schmidt number of 660) as a function of the 10 m neutral wind speed during a) winter and b) summer. The dots represent the half-hourly values of k_{660}. The color represents the water-side convective scale ($w*$) for data under unstable atmospheric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

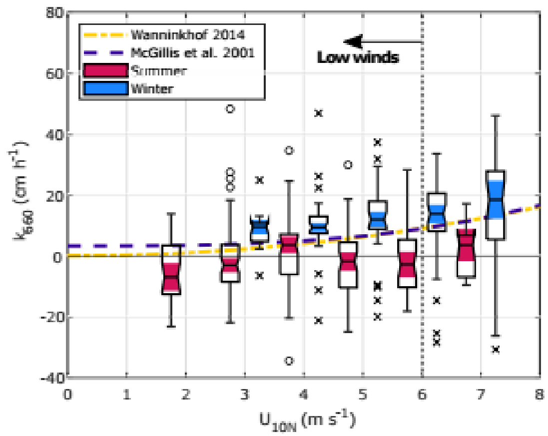

Figure 10. Boxplots of the gas transfer velocity for CO_2 (adjusted to a Schmidt number of 660) during unstable atmospheric conditions as a function of the 10 m neutral wind speed during summer (pink) and winter (blue). The median, first, and third quartiles are represented in each box; the whiskers 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.

***Appendix A** with the full set of figures used for the analysis of the normalized gas transfer velocity. An example of such a figure is included below (**Figure A1**) presenting the water-side controls during high, intermediate, and low wind speed conditions. Similar figures (**Figure A2** and **Figure A3,** not included here) show the corresponding results for the wave field and atmospheric controls for the three wind speed regimes. Additionally, **Figures A1** to **A3** include the probability distribution of each parameter for the three wind speed regimes, this to highlight the differences between wind-speed regimes.*

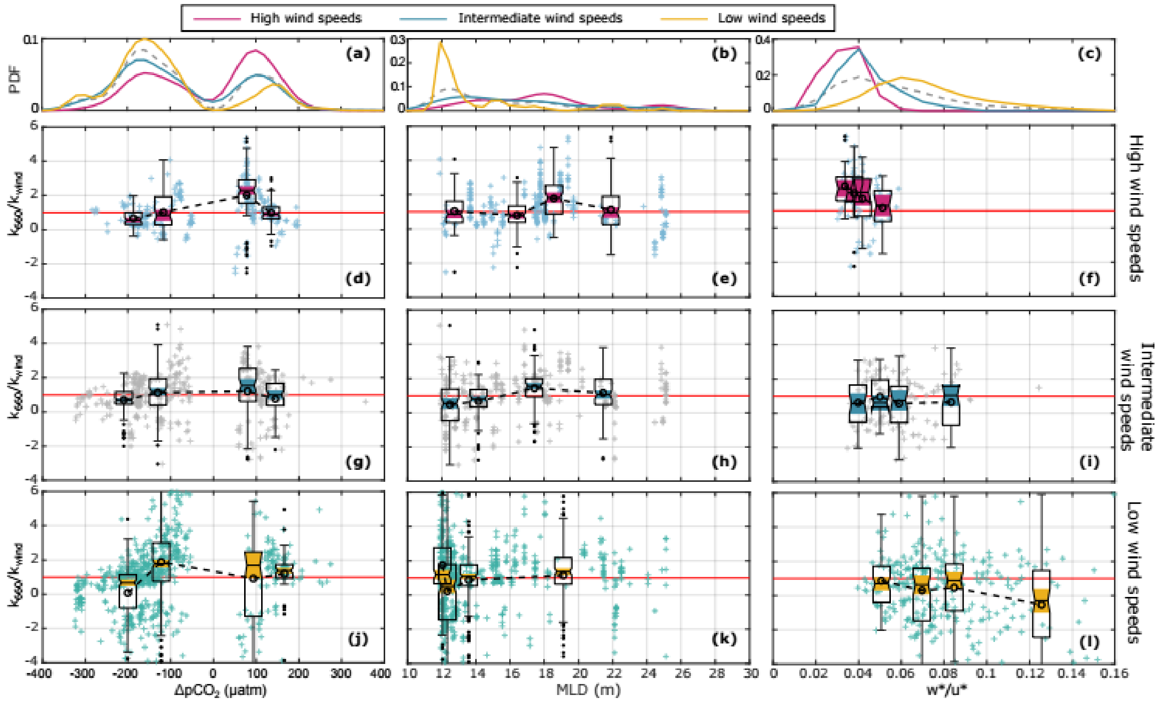

Figure A1. Water-side control mechanisms: ΔpCO_2 (left), mixed layer depth (center), and water-side convective scale normalized with the friction velocity ($w*/u*$) (right). Top panels a),b), and c) show the probability distribution function (PDF). Panels d) to l) show the normalized gas transfer velocity (k_{660}/k_{wind}) under high (upper), intermediate (middle) and low wind speeds (lower). 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. 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 $k_{660}/k_{wind} = 1$.

Secondly, it is not clear what benefit was gained by working with such a long (nearly a decade) EC dataset, as the correlation-based analysis of GL is similar to those applied to shorter-term datasets from the same measurement platform. I understand that a detailed time-series analysis was beyond the scope of this work, but a short discussion may be useful. So, maybe the authors can offer some advice as to the time required to capture the full range in gas transfer variability? i.e., for readers planning a similar coastal EC deployment, is it enough to measure for a year, or do we need many years to capture the variation in physical forcing described in GL?

The EC methodology can be very convenient for flux analysis; however, the amount of data that often has to be discarded due to quality control can become a major issue. In the case of this study, some of the quality control criterion were very strict, which lead to a high percentage of rejection. Particularly, the fact that we used data only from the open-sea sector caused the rejection of 85% of the data. In other studies, depending on the objectives of the work, the characteristics of the study site, and the set-up of the platform, it might be possible to relax some of these criteria. Thus, keeping a larger proportion of the data.

Furthermore, we were interested in capturing the short-term variability in order to associate the local processes to the gas exchange, as well as the long(er)-term variability to assess the seasonal and inter-annual patterns. To this aim, a significantly longer record was required compared to previous studies from the same site. Some of these studies had the objective to evaluate the effect of single processes on the gas exchange (e.g. Rutgersson and Smedman, 2010, Norman et al., 2013).

The first paragraph of the ***discussion*** *was modified:*

"We used nine years of eddy-covariance-based FCO_2 data to evaluate the effect of different control mechanisms on air-sea CO_2 gas exchange. By using this long record, we were able to capture the seasonal and inter-annual variability of the FCO_2 and other parameters relevant to the gas exchange (Sect. 3.1), as well as directly assess controls on k_660 (Sect. 3.2). These long records of direct FCO_2 measurements are not common, and often not necessary when evaluating local effects of single processes on air-sea gas exchange. However, continuous measurements over long periods are necessary in other to resolve the effect of multiple parameters on the gas exchange at both short- and long-term (several years) scales."

- **Line-by-line comments:**

92: I see that instruments were located 9m above the ground surface, but how high is this above the sea surface?

The base of the tower is at 1.4 m above the msl (Sjöblom and Smedman, 200). Thus, the height of the instruments is 9+1.4=10.4 m above the msl.

The sentence "(i.e. 10.4 m with respect to the mean sea level)" *was included.*

98: Was z/L uniform across wind directions within the southeast window?

The distribution of z/L across wind directions (from southeast, i.e. open-sea sector) was relatively uniform (see figure RC1.1 below), in particular, for the neutral and stable conditions. For unstable conditions, there was a larger number of cases from the more eastward directions. However, we do not think this fact would have major implications in our analysis.

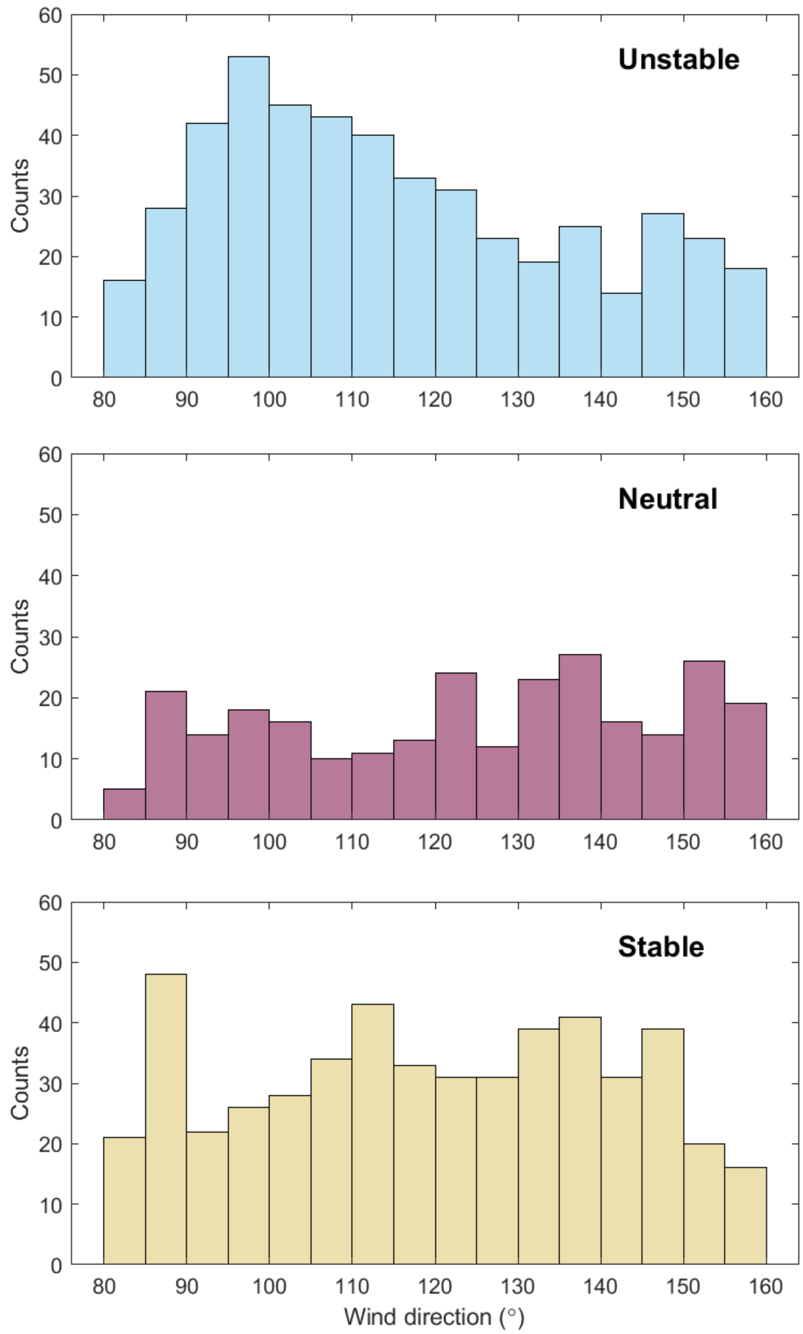

Figure RC1.1. Histogram showing the distribution of the atmospheric stability (z/L) across the wind directions corresponding to the open-sea sector. Only cases where both FCO2 and k660 data were available were included in the figure.

No further changes were made in the manuscript.

110: Are there ancillary measurements of T (e.g. from a shaded thermometer, or maybe a closed-path IRGA) to show whether or not solar heating of the sonic anemometer affected the Ts record?

There are additional measurements of temperature at different levels in the tower (from a profile measurement array). The profile air temperature observations are carried out continuously with mechanically-ventilated air temperature sensors. Some comparisons have been made between the sonic temperature data and the profile data (not directly as part of this work). The analysis has shown that major differences occur between these observations during precipitation and heavy fog events due to disturbances in the sonic anemometer. However, no major effects have been observed due to solar heating.

In general, we would not expect a major effect of solar heating on the air temperature measurements from the sonic anemometer as these are obtained from the determined speed of sound. Furthermore, only the turbulent fluctuations of the sonic temperature were used in the flux calculations; these fluctuations would be even less susceptible to disturbances caused by solar heating of the sensor. Finally, we consider that some of the criteria used as part of the quality control process would, in any case, remove low quality data from the sonic anemometer. For example, removing data at very low wind speeds (when solar heating would be expected to be significant) and precipitation events (based on the RSSI of the gas analyzer and relative humidity). Thus, ensuring that only good quality and undisturbed data is used in the analysis.

No further changes were made in the manuscript.

Data processing: I would like to see more detailed statistics showing how many 30-min datapoints were rejected according to individual screening criteria. Comparing, for example, figure 5 with the full time-series in figure 4, it appears that a large majority of data failed the screening criteria. If so, this needs to be fully explained in the methods.

Thanks for pointing this out. Yes, a very large percentage of the data was removed during the quality control processing. In particular, restricting the analysis to the open-sea sector removed ca 85% of the initial data set. This, in addition to other quality control steps which also had an effect on the final size of the data set.

The final data set consisted of 18.7% of the initial FCO2 data and 15% of the k660 data, with respect to the total amount of data available for the open-sea sector. When comparing with the total available data (i.e. all wind directions) this percentages go down to 2.3% and 2.0%, respectively.

In ***Data Processing section*** *(2.2) the following paragraph was included:*

"The final data set, after quality control processing, consisted of 3,477 FCO_2 data points and 1,349 k_660 data points. This amount of data corresponds to 18.7% and 15%, respectively, out of the total amount of data available for the open-sea sector (18,625 FCO_2 and 8,974 k_660 data points). Further information about the rejection rates of each quality control criterion is presented in Appendix B."

***Appendix B** was included where a more detail 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
$\lvert FCO_2\rvert_{min} = 0.05\,\mathrm{\mu mol\,m^{-2}\,s^{-1}}$	51.4	48.4
$\lvert\Delta pCO_2\rvert_{min} = 50\,\mathrm{\mu atm}$	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.

174: The generation of excessive negative k values is a frequent criticism, and major caveat, of EC-based gas transfer studies. So, what is the justification for removing -k values when they otherwise meet the screening criteria applied to the rest of the dataset? Doesn't removing negative values artificially decrease the variability in calculated k?

We agree with this comment, removing the negative k values can indeed create a bias in the analysis. We have initially thought that these negative k values would introduce some unrealistic results as we could not find a feasible explanation for them, even if they had fulfilled all quality control steps. However, we have reconsidered this earlier decision and negative k values are now included in order to avoid a bias in the data and the subsequent analysis.

*All data fulfilling the quality control procedures were considered in the analysis, including the negative k_660 values. The corresponding modifications were made throughout the manuscript. In particular, modification were made to the data processing **section 2.2** and in the results from **section 3.2** and onwards.*

177: As per the discussion above, I do not agree that k_r "remov[es] the wind-speed dependency from k_660". Given that the majority of the analysis in GL revolves around k_r, I think some additional description of the W14 parameterization and it's applicability to the current study site is warranted.

In the revised manuscript, the concept of a residual gas transfer velocity (k_r) as described in the initial version of the manuscript is no longer used. Therefore, we consider that further *description and discussion of the W14 (or Mc01) parameterization(s) is no longer needed. However, these parameterization were still included as a reference in Figure 5, for example, to put our own observations in context to other commonly-used relationships.*

245: This enhanced wind dependence of k under unstable conditions is consistent with prior work in the Baltic (https://doi.org/10.1007/s10546-018-0408-9) and elsewhere (https://doi.org/10.1007/s10546-018-0408-9; https://doi.org/10.1002/lno.11620). Since these conditions are associated with the largest deviation of measured k_660 from k_W14, can the authors offer any further ideas as to the major driving cause?

The enhanced gas transfer velocities under unstable atmospheric conditions might be associated with an increased small-scale turbulence, which in turn can modify the characteristics of the ocean surface as suggested in Andersson et al., 2018, among others. The results of this study, however, present further evidence showing that the enhanced transfer was caused by a combination of conditions (including unstable atmospheric stability). We therefore suggest that sea spray during such particular conditions is the mechanism enhancing the positive fluxes at intermediate and high wind speeds, and not the individual effect of a single parameter such as atmospheric stability.

A recommendation to investigate further the effect of sea spray on air-sea CO_2 gas exchange was included in the ***discussion*** *and* ***conclusions.*** *Additionally, a sentence stating the potential relevance of atmospheric processes on CO_2 transport was included in the* ***discussion.***

259: Couldn't the lack of relationship between the wave field and k_r be in part explained by the fact that (as explained by the authors), the waves here are not swell but rather locally-generated by wind? I.e., one would expect a strong correlation between wind and wave height here?

Definitely. Under the observed conditions, the wind and waves were strongly correlated (see Figure RC1.2 below). We can expect a larger proportion of swell at the lower wind speeds (0-2 m/s), however, these data were removed as part of the quality control.

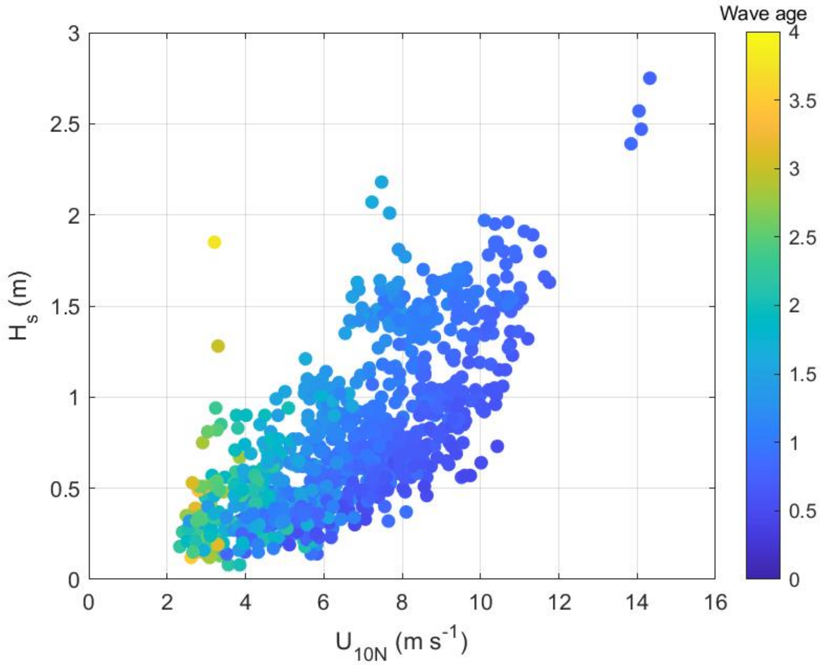

Figure RC1.2. Significant wave height vs neutral 10-m wind speed. The color represents the wave age (Cp/U_10N). The data corresponds to the open-sea sector conditions. Only data points with available FCO_2 and k_660 were included in the figure (i.e. data used in the k660 analysis).

299: Maybe I missed it, but I do not see where the formulation of McGillis 2001 is compared with the k values calculated by GL.

The formulation of McGillis 2001 (Mc01) was only included to provide a visual reference of a cubic relationship between k_660 and U10N (i.e. Figure 5). In the revised version of the manuscript the use of a residual gas transfer velocity calculated using a wind-based parameterization is no longer used. Thus, we consider that a detailed comparison of our data set with Mc01, or other parameterizations is beyond the scope of this study.

References:

Andersson, A., Sjöblom, A., Sahlée, E., Falck, E., & Rutgersson, A. (2019). Enhanced air–sea exchange of heat and carbon dioxide over a high Arctic Fjord during unstable very-close-to-neutral conditions. *Boundary-Layer Meteorology*, 170(3), 471-488.

Norman, M., Parampil, S. R., Rutgersson, A., and Sahlée, E. (2013). Influence of coastal upwelling on the air–sea gas exchange of CO2 in a Baltic Sea Basin. *Tellus B: Chemical and Physical Meteorology*, 65(1), 21831.

Rutgersson, A., and Smedman, A. (2010). Enhanced air–sea CO2 transfer due to water-side convection. *Journal of Marine systems*, 80(1-2), 125-134.

Sjöblom, A. and Smedman, A.-S.: The turbulent kinetic energy budget in the marine atmospheric surface layer, *Journal of Geophysical Research: Oceans*, 107 (6–1), 2002.

Van Dam, B. R., Lopes, C. C., Polsenaere, P., Price, R. M., Rutgersson, A., & Fourqurean, J. W. (2021). Water temperature control on CO2 flux and evaporation over a subtropical seagrass meadow revealed by atmospheric eddy covariance. *Limnology and Oceanography*, 66(2), 510-527.