Response to John Prytherch's comment

General comments:

RC: This paper presents air-sea CO2 flux measurements made from a research vessel in the Southern Ocean. The flux measurements were made using four Licor infra-red gas analysers, two of which used a dried airstream in order to minimise sources of error. The measurements from the other "wet airstream" sensors had an additional correction applied to remove the effects of humidity cross talk using a published method (the PKT correction, Prytherch et al., 2010a). The results obtained using the PKT correction were in poor agreement with those from the "dried airstream" sensors. The authors then perform an analysis of the PKT correction method, and demonstrate that it does not properly correct CO2 fluxes. At the time of publication in 2010, the PKT correction method was the only one available for open-path sensors. These sensors were used on platforms where it was not practical to dry the airstream (e.g. on buoys). The method has since been used, and the results published, by various research groups. Therefore, the authors refutation of the PKT method is an important result and should be published. The authors use a mathematical analysis of the PKT method to demonstrate that the method is invalid. This important result should be made as convincingly as possible. While the analysis itself is well constructed, we feel that the presentation of the results could be improved. We hope that our suggestions below will help improve the clarity and impact of the paper.

AC : We would like to thank John Prytherch and Margaret Yelland for carefully reviewing our submission without prejudice and for providing detailed and extremely helpful comments and suggestions.

Specific comments:

RC: a) It is not clear from the bottom panel of Figure 3 that Eq. (7) obtains similar results to the "standard" PKT correction. We compared Eq. (7) and the standard PKT results from our own air-sea CO2 flux data (previously published in Prytherch et al., 2010b), and obtained a clear 1:1 relationship (R2 = 0.98), shown in the attached Figure R1 (Measurements in all figures are from the HiWASE field campaign with quality control as described by Prytherch et al. (2010b)). The addition of a similar plot from the authors data would be much more convincing than the current time series shown in Fig. 3.

AC : One intention for showing the results of Eq. (7) in Fig. 3 was to show that Eq. (7) and the PKT results are identical. We agree that it is not clear from Fig. 3 only. However, in section 4 we show that PKT and Eq. (7) are mathematically identical. The second intention was to show the results that were rejected by the PKT correction. However for additional clarity we have explicitly plotted the results of Eq. (7) against the PKT corrected results in Fig. 9 (please note that the figure numbers have changed based on the original submission).

The large scatter in Fig. R1 is surprising, since PKT results and Eq. (7) are identical, see Fig. 1. There should be no scatter, but a 1 : 1 relation.

We would like to ask, how Eq. (7) was applied to the HiWASE data set? Was the factor β set to 2, or calculated for each flux interval?

RC: b) The authors analysis of the PKT correction shows that the "corrected" flux value is a product of a detrended (with respect to humidity) flux and a "beta" term. The beta term includes a factor of 0.5, which comes from a term in the original iterative PKT correction (Eq. 6), which was used to help the iteration converge by making the individual steps in the iteration smaller. This arbitrary factor was not thought to affect the convergence value, but the authors show that in fact the PKT correction is directly dependent on the value of this factor. Modifying this value in the iterative PKT correction would provide a convincing validation of Eq. (7) (and hence the invalidity of the PKT correction). We have done this using our flux data, using a factor or 0.75 rather than 0.5 in the original PKT correction. This changes the value of beta from ≈ 2 to ≈ 4. The attached Figure R2 shows that the "PKT 4" results are approximately equal to 4 times the detrended flux.

Figure R3 shows that if this factor is used in the sensible heat flux test, then the con-verged values of the fluxes are twice as large as the actual sensible heat flux. It was a very unfortunate coincidence that the detrended sonic temperature flux was a factor of 2 smaller than the sensible heat flux, and that the factor of 0.5 used in the iteration happened to increase the detrended flux by a factor of 2.

AC : Fig. 8 (Now Fig. 10) was modified following the suggestion made in Fig. R3 to show the dependence of the PKT correction on the iteration step width. A paragraph was also added to the end of Sect. 4 to point out the connection between β and the iteration step width in Eq. (6).

RC: c) Paper structure. The main (or only) result in this paper is the proof that the PKT method is not valid (which begs a minor question - why was this paper not submitted to Geophysical Research Letters as a "comment"?). However, the narrative structure used (ie "we made these measurements, then we tried this correction. The correction didnt work on our measurements, so we examined it and found it to be incorrect") leads to the authors giving a very detailed description of the lack of agreement in the results between the data from the dried sensors and those from the PKT-corrected wet-airstream sensors. Given that the subsequent analysis demonstrates that the PKT correction does not work, much of this description seems superfluous.

AC : We agree that the mathematical proof of Eq. (7) does not require experimental validation. We believe, however, that the presentation of real measurements will increase the impact of this submission. Apart from the refutation of the PKT correction, we also show that the humidity crosstalk-related bias can affect the closed-path analyser when the air is neither dried nor filtered. We also show that the application of membrane dryers does not alter the CO_2 flux measurements, see Figure 6 (was Fig. 5). We added two sentences to the conclusion to underline those additional findings.

Technical Corrections:

RC: Is there a reason why the two open-path 7500 units were chosen to be the "dry" sensors whereas the two closed-path 7200 sensors were chosen to be "wet"? The terminology of "open" and "closed" *is used rather loosely - the 7500 is an open path sensor, but in the SOAP experiment it was effectively converted into a closed path sensor. This could be explained more clearly, e.g. was the conversion to closed path done using the method of Miller et al., 2009?*

AC : We decided to use the 7200 unit as "wet" sensor, assuming that the build-in pressure and temperature sensors would allow a high standard air-density correction and make the drying of the air-stream superfluous. We did not expect to observe similar biases as for open-path deployments.

The conversion of the 7500 open-path units was as in Miller et al., 2010. This is described in Page 28285, line 14ff, however the sentence structure was not clear, for which we apologize. This section was rewritten to clarify.

RC: Page 28286, line 8. How was the conversion to CO2 mixing ratio done? Was it done in a similar fashion to Miller et al., 2010?

AC : The mixing ratio was calculated as the ratio of measured gas density to the dry air density, which was calculated using the ideal gas law and accounting for the measured water vapour concentration. This is described in the introduction on Page 28282 lines 1-5. This is equivalent to the conversion presented in Miller et al., 2010. A cross reference was added to Page 28286, line 8 to point this out.

RC: Page 28287, line 7 states that no airflow distortion correction was applied, but the caption for Figure 2 states that wind speed was corrected for airflow distortion. This could be confusing; perhaps specifying that "[Flux] measurements were not corrected: : :" would clarify this.

AC : "flux" was inserted in Page 28287, line 7 to clarify.

RC: Page 28288, line 2. Results from two wet IRGA are shown, so plural "analysers".

Page 28289, line 11. An erroneous "the" after ": : :scatter, but: : : ".

Table 1. Prytherch et al., 2010b presents PKT corrected results and should be added to this list.

AC : Thanks for pointing these mistakes out. Changes are made to the text and table 1 according to the suggestions.

RC: Page 28288 line 7. Could the authors comment on potential reasons why the range of bias was so much larger for IRGAwetA than wetB?

AC : We do not understand the true nature of the bias at the current stage.

RC: Page 28292, line 5. The fact that the bias between the "sonic flux" and the sensible heat flux is *small is not really relevant: the intention of the original test was to see if the detrended flux (with a large bias) was iterated back to the correct value or not. Unfortunately it was...*

AC : We agree and will remove this sentence.

RC: Figures. The general presentation of the figures needs to be improved. Some examples are listed here: Missing "-" signs in the exponents in Figures 2 to 5. Figures 8 and 9 use W/m2 rather than W m-2 as used in the other plots. Labels use wetA, dryB etc but in the figure captions and text IRGAwetA etc is used. Inconsistent use of capitalization (e.g legend of Figure 9). Missing space in x-axis label of Figure 9. "w" should be capitalized in the colour bar label of Figure 6. Labels outside legends in Figures 8 and 9.

AC : The "-" signs in all figures except in 1,6 and 7 were lost during the type setting process, when the eps files were converted to pdf. We apologize for having overlooked this important detail. Figures with correct signs are provided in a separate comment. The inconsistencies and capitalization errors will be removed. Again thanks for pointing these out!

RC: Figure 2. What bulk formulae were used to calculate the bulk fluxes? Also, shaded areas are shown in this figure, but are only explained later on in the caption to Figure 3.

AC : The TOGA COARE 3.0 algorithm was used to calculate the bulk fluxes. The label in Figure 3 will be changed to reflect this.

RC: Figures 6 and 7. I wondered if some of the difference between the binned WetA and WetB results might be due to data from different periods being binned. The total number of periods from each of the two sensors only differs by 4 (174 for A, 178 for B), but it is not clear that the majority of these data come from the same periods, rather than data from WetA being accepted for one period, and WetB from another.

AC : The majority of the data come from 161 intervals where both *wetA* and *wetB* data was accepted by the PKT quality control. Restricting the bin averaging to this 161 intervals does not significantly change the the slopes and offsets in Figures 6 and 7. Figures 6 and 7 (now figures 7 and 8) will be updated to show only the shared 161 measurements. The general quality control was passed by both sensors wetA and wetB for 267 intervals. This information will be added to the text.

We want to point out here, that table 2 had been updated. During the preparation of the manuscript we had mad our quality control more restrictive. This lead to the removal of additional 30 data points. However, we forgot to update table 2 in the document before submitting it to ACPD. The new table 2 is based on 161 total data points (191 in the old version). The computed average values did change. This has as well impact on the numbers in Sect. 3.2. The conclusion of this work is however not changed. We apologize for this mistake.

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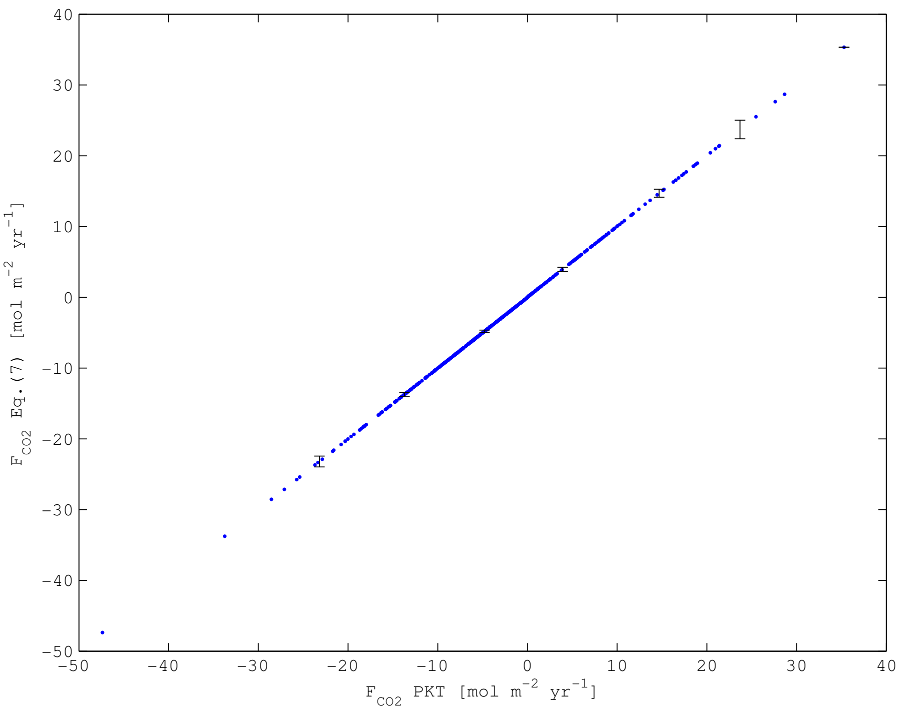

Figure 1: (Fig. 9 in the revised manuscript) Scatter plot of the results from Eq. (7) against the results of the PKT correction for IRGA *wetB*, individual (blue) and binned (black).

Response to Mingxi Yang's comment

General comments:

RC: This paper describes a recent set of air-sea CO2 eddy covariance flux measurements. Four effectively closed-path CO2 sensors were used, two dried to minimize the bias due to H2O cross-correlation, two undried (i.e. sampling moist air). Using flux from the dried sensors as reference, the authors convincingly demonstrated that the correction scheme based on similarity theory (i.e. PKT correction) does not remove the bias in the measured CO2 flux from the undried sensors under conditions of large latent heat flux.

The direct measurement of air-sea CO2 flux is obviously essential for the understanding of global carbon cycling and climate, with the open path sensors (e.g. Licor 7500) widely used. Since its publication (Prytherch et al. 2010a), the PKT method has been tried by several authors to correct eddy covariance CO2 flux. Thus it is important for the paper under consideration to be published.

The content of the paper is sound. Moreover, Referee #1 (who published the PKT method) already agreed with the authors of this paper. I only have a few anecdotal and editorial comments below:

AC : We wish to thank Mingxi Yang for his attention to our submission and the provided comments and suggestions that helped us to advance the publication.

RC: 1) The authors missed an opportunity to strongly recommend the drying of IRGA sensors, which appears to be the most reliable method thus far for making CO2 measurements.

AC : We appreciate the suggestion and added a sentence to the conclusion recommending the Miller et al (2010) method.

RC: 2) How does H2O cross-contaminate the CO2 flux? Its probably not related to sea salt, since the 7200 sensors are operated inline during this experiment. Knowing the cause for this cross-contamination might lead to improvement in open-path CO2 sensors. Any educated guesses?

AC : We are aware of the importance of this problem for the air-sea gas-exchange community and will continue searching for a solution. However at the current stage we have no educated guess.

RC: 3) This cross-contamination presumably cannot be clearly identified in the cospectrum. The authors can mention that spectral analysis by itself is inadequate as a quality control filter for CO2 fluxes.

AC : The following sentence was added to Sect. 2.1 "It has to be noted here, that the co-spectra of CO2 and H2O are similar. Therefore a cross-contamination of the CO2 signal with H2O can not be clearly identified by spectral analysis."

RC: 4) For the gas exchange community, it would be insightful for the authors to obtain the data from Prytherch et al. 2010b and see what the k values from HiWASE are like only for conditions of near-zero latent heat flux.

AC : The scope of this contribution is to show that the PKT correction is not as successful as initially considered, and that the "closed path with diffusion dryer" method is the only reliable one for making EC flux measurements of CO_2 over the ocean with Licor sensors. We would find it more appropriate for the authors of Prytherch et al. (2010b) to re-publish the HiWASE data and derived k values. However, it should be noted that for this study, the chosen latent heat flux limit of 7 Wm^{-2} did restrict the wind speed range to 11 ms^{-1}.

Specifics:

RC: Another recent paper that utilized the PKT correction (and suggested that it did not work) is Ikawa et al. (2013) (www.biogeosciences.net/10/4419/2013/).

AC : Thanks! We added a reference to Ikawa et al. (2013) and Huang et al. (2012) in the introduction and to table 1.

P 28282, line 18. Rather than "restricted to", it's more accurate to says "the EC method provides relatively robust CO2 flux measurements (uncertainty of $\sim$ %) in regions with air-sea gradient. . . "

AC : The text was changed following the suggestion.

RC: P 28283, line 9. The point of having a very high flow rate is to maintain a fully turbulent flow. Would be more insightful to present the critical Reynolds number here in addition to the number of SLPM.

AC : We added the critical Reynolds number of 2100 here and also give an estimate of the Reynolds number for the flow in the inlet tubing in Section 2

RC: 28285, line 9. What temperature was the inlet heated to, in order to preserve the latent heat flux? Water vapour is well known to be sticky. Thus even in the absence of condensation, significant attenuation of water vapour flux at high frequencies is possible.

AC : The inlet tubing temperature was not recorded continuously, but the LICOR temperature sensors recorded temperatures ranging from 23°C to 36°C at an outside air temperature ranging from 8°C to 16°C. We cannot ensure that the water vapour flux was fully resolved by the un-dried sensors. However this does not affect the CO2 flux measurement, since the air-density flux correction is done with the humidity fluctuations in the measurement volume.

RC: 28287, line 3. The authors havent showed that the IRGAdry measurements are completely unbiased. For example, was 97% of the H2O removed by the drier, as in Miller et al. (2010)? Also, *is there any residual contribution of sensible heat flux to IRGAdry?*

AC : We added Fig. 1 to the manuscript to show a time series of the air-density bias fluxes in comparison with the CO2 flux measured by the IRGA *dry*. The application of the diffusion dryer reduced F_q in average by 93%, from 36Wm^{-2} average latent heat flux magnitude to 2.4Wm^{-2}. All four IRGAs do also need a non-zero correction for sensible heat flux and the cross-correlation of pressure and vertical wind speed. We did however not find a correlation between the pressure or temperature bias fluxes and differences between the final CO2 flux estimates. As far as our observations go, the problem seams to be solely with H2O.

RC: P 28288, line 2. The Wanninkhof (1992) parameterization is now widely accepted to be too high due to a bias in the global radiocarbon estimate. This should be acknowledged if cited. If the authors believe k to be a quadratic function of wind speed, the Sweeney et al. (2007) parameterization would seem more appropriate.

AC : We do not favour any parameterization but decided on picking one that is widely used in the gas-exchange community, to put our CO2 flux measurements in context. However, as you mention Sweeney et al. (2007) have shown that Wanninkhof (1992) needs to be corrected, we updated the parameterization used in Fig. 3.

RC: Line 8. Why did IRGAwetA give much larger scatter than wetB?

AC : This is not clear to us, but we assume that understanding why the two IRGA did behave different will help understanding the bias and mabye correcting for it.

RC: P 28290, line 4. To investigate the unsatisfactory. . . instead of in the light of the unsatisfactory. . .

AC : This sounds better, thanks.

RC: P 28290, line 15, parenthesize 0 in xc0 to be more consistent

AC : Did this; thanks

RC: p 28292, line 11. definition of F0TS? Line 21. Overestimation of CO2 flux magnitude

AC : The definition was added and the word "magnitude" added to the text.

RC: P 28294, line 3. As this is the summary section, rather than using nomenclatures, you can simply say that the PKT correction applied to undried IRGAs reduced the scatter but did not reduce the bias in flux compared to dried IRGAS. Line 11. . . .to retrieve the true CO2 flux from. . .

AC : We removed the nomenclature from the conclusion and the word ”true” inserted. It reads easier now, thanks!

RC: Fig 3 Legend flux calculated based on the parameterization of. . .

AC : This was changed.

RC: Fig 4. Legend Difference between. . .

AC : This makes more sense.

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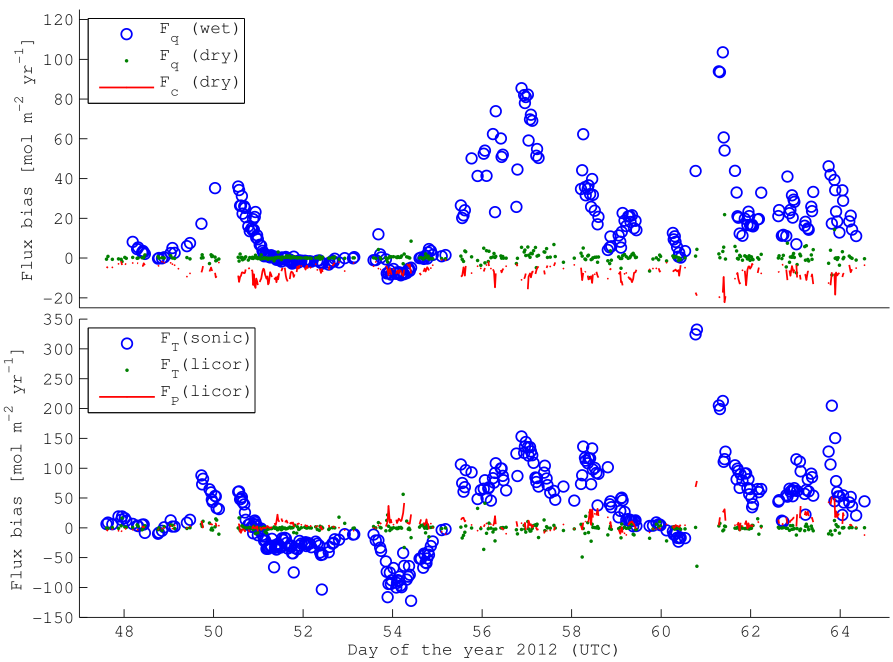

Figure 1: (Fig. 4 in the revised manuscript) Time series of the bias fluxes in Eq. 3, caused by air density fluctuations (Webb et al., 1980). Top: Bias flux caused by humidity fluctuations F_q upstream of the dryer *wet* and down stream *dry* and the CO2 flux F_c as measured by the IRGA *dry* (there are only small differences between *A* and *B*). Bottom: Bias flux caused by temperature fluctuations F_T as measured by the bow mast sonic and as measured by the CP-IRGAs, and the bias flux caused by pressure fluctuations F_P.