Authors' response to 'Reviewer comment for os-2018-108' by E.S. Saltzman

We would like to express our gratitude to E.S. Saltzman for his thorough and helpful review. Our answers are given in the table below.

Reviewer's comment	Authors' response
There seems to be a rather strong bias against eddy covariance in this paper - the only comment about more than a decade of new work in that area is rather dismissive and citation-less. So, a reader new to the field would imagine that eddy covariance is not generating insight into air-sea gas transfer (which I think most would agree is not the case). The fact that eddy covariance data are often binned seems like an odd criticism, especially when the dual tracer method (which requires long averaging) is held up as the "gold standard". The uncertainty in a single DMS eddy covariance measurement under favorable conditions is on the order of 25% and one could easily imagine interesting results from simultaneous eddy covariance and active thermography measurements.	It was in no way our intention to proclaim the dual tracer technique to be the 'gold standard'. To put our measured gas transfer velocities into perspective, we chose to compare them with the Ho. et al. 2011 parameterization. We decided against using more than this one parameterization, since the many available parameterizations for the transfer of CO_2 for both the dual tracer technique and the eddy covariance technique are very similar in the range of wind speeds we studied. The Ho. et al. 2011 parameterization was chosen because it is one of a few parameterizations where a confidence interval is given. Our reasoning would not change if we used a parameterization based on eddy covariance of CO_2 to compare our data with. We have changed the manuscript to include an eddy covariance based CO2-transfer parametrization to Fig. 6 and also included some eddy covariance DMS measurements. We have extended the discussion in the results section to also discuss the mentioned additions to Fig. 6. We mentioned that binning is commonly done for eddy covariance data sets to put the temporal resolution of the ACFT into perspective, not to criticize eddy covariance. We agree that simultaneous measurements of eddy covariance and ACFT would be very valuable.
I was surprised that so much emphasis in this paper was placed on the dual tracer method because active thermography captures only interfacial flux. Bubble-related transfer is very important for CO2 so if active thermography agrees with dual tracer at intermediate and high winds, then it would seem that some assumption in the interpretation of these methods is wrong. Active thermography should be more similar to eddy covariance measurements of DMS than to a dual tracer fit meant to mimic CO2. Eddy covariance studies of DMS and CO2 clearly show that CO2 fluxes at intermediate and high winds are enhanced by bubble transfer relative to dms (which is controlled mostly by the interfacial flux; for example, Bell et al., 2013; Blomquist et al., 2017). There is a conundrum here - if the dual tracer method gets kco2 right (which it seems to), it must be bubble-enhanced also. So one would expect active thermography to diverge from the dual tracer results at intermediate and higher wind speeds.	Iwano et al. 2013 (CO2, solubility 0.8) and Krall&Jähne2014 (two tracers with solubility 1 and 3.2) found the measured gas transfer velocities to be compatible with the theoretical prediction for pure interfacial transfer of $\mathrm{k} = \beta^{-1} \mathrm{u}_{*} \mathrm{Sc}^{-\mathrm{n}}$ up to wind speeds of around 30-35 m/s. Both of these studies were done in a wind-wave tank using fresh water, in which the bubble size distribution differs from sea water. However, gas transfer velocities measured in a hurricane (McNeil&D'Asaro2006, O2 with solubility of 0.03) agree well with the transfer velocities measured in both lab studies mentioned above. From this line of evidence we can infer that bubble mediated gas transfer is weak for winds up to 30-35 m/s for gases of most solubilities for fresh water and for sea water. We therefore disagree with the assertion that "bubble-related gas transfer is very important for CO2" for winds lower than 30-35 m/s. In addition, Nagel et al. 2015 found no differences between simultaneous heat transfer and gas transfer measurements for wind speeds up to 12.7m/s, indicating that bubble enhancement for gas transfer is not significant at those wind speeds. Thus we think that the differences found between EC measurements of CO2 and DMS must have causes other than bubbles.

Several studies suggest that interfacial gas transfer appears to be limited at higher winds. This is attributed to wave shielding and other wave-related effects demonstrated in the laboratory by Mueller and Veron (2009) and incorporated into gas transfer models by Fairall et al., 2011 and Donelan and Soloviev, 2016. Such processes could be salient here in relating active thermography to gas transfer. This is not to say that the arguments in the paper about fetch and surfactants etc. are not very well founded. I think they are.	It is true that several studies found a limitation of the Drag coefficient at higher wind speeds (Mueller and Veron 2009, Takagaki et al. 2012). However, the gas transfer velocity has no such limit, see the studies referenced above (Iwano2013, Krall&Jähne2014, McNeil&D'Asaro2006). Since, as argued above, bubble effects are weak up to 30-35m/s, we can assume that transfer velocities measured at wind speeds below 30-35 m/s are controlled by the transfer through the water surface, and are independent of the gas or measurement technique (EC or DT or ACFT) used.
But the overall premise that the dual tracer and active thermography measurements should measure the same thing seems open to debate. I think this should be considered by the authors and perhaps addressed in the manuscript.	Simultaneous measurements of heat transfer and gas transfer with a mass balance method have shown that heat transfer velocities can be scaled to gas transfer velocities for wind speeds up to at least 12.7 m/s (Nagel et al. 2015). No evidence of bubble contribution was found in the measured gas transfer velocities. Therefore, we think that a comparisons with a dual tracer parameterizations is valid.
	Changes to the manuscript: we have extended our reasoning why we think that surfactants are the most likely cause for lower gas transfer in the Aranda2010 campaign. We discuss why we think that bubbles do not explain the lower gas transfer using the arguments given above.
	references: Iwano et al. 2013: Mass transfer velocity across the breaking air–water interface at extremely high wind speeds https://doi.org/10.3402/tellusb.v65i0.21341 Krall & Jähne 2013: First laboratory study of air–sea gas exchange at hurricane wind speeds https://doi.org/10.5194/os-10-257-2014 McNeil & D'Asaro 2006: Parameterization of air–sea gas fluxes at extreme wind speeds https://doi.org/10.1016/j.jmarsys.2006.05.013 Mueller & Veron 2009: Nonlinear Formulation of the Bulk Surface Stress over Breaking Waves: Feedback Mechanisms from Air-flow Separation https://doi.org/10.1007/s10546-008-9334-6 Takagaki et al. 2012: Strong correlation between the drag coefficient and the shape of the wind sea spectrum over a broad range of wind speeds https://doi.org/10.1029/2012GL053988 Nagel et al. 2015: Comparative heat and gas exchange measurements in the Heidelberg Aeolotron, a large annular wind-wave tank https://doi.org/10.5194/os-11-111-2015