Review of: ACP-2017-85 - "**Estimation of bubbled mediated air/sea gas exchange from concurrent DMS and CO2 transfer velocities at intermediate-high wind speeds" by T. G. Bell et al.**

Reviewed by Ian Brooks

This paper presents direct measurements of the air-sea fluxes and transfer velocities of both CO_2 and DMS from a cruise in the North Atlantic. The difference in behaviour of the transfer velocities as functions of wind speed has been widely assumed to be a result of the different contributions of bubble mediated fluxes for gases of very different solubility. Here, simultaneous measurement of both gas fluxes allows a direct estimate of the bubble mediated flux to be calculated for open ocean conditions, and a comparison to model predictions to be made.

This is an important contribution to the field, and deserving of publication after revision for, mostly minor, deficiencies detailed below.

DETAILED COMMENTS:

Line 86-87: "In that study, no statistically significant difference was observed in gas transfer-wind speed relationships of CO2 and DMS for winds below 10 m s^{-1}" – need to clarify this statement, was a significant difference found for winds above 10 m s^{-1}? Was 10 m s^{-1} the maximum wind speed in the study?

Line 104-105: as noted in the comment from Blomquist, the air-side resistance is not a function of solubility (though its contribution to the derivation of waterside transfer velocity is dependent on solubility).

Line 122-123: The use of the COAREG 3.1 model to calculate air-side transfer velocities in order to derive the waterside transfer velocity introduces an assumption that COAREG is providing valid values of k_a. Any uncertainty in this will impact the later results and should be acknowledged and if possible quantified.

Line 126-127: "The air side gas <u>transfer</u> contributes about 5% on average to the total resistance for DMS" – do you mean 'air-side resistance' rather than air-side transfer?

Line 140: "...(Equation 4)..." should be "...(Equation 3)..."

Line 170: "...into Equation 6 yields..." should be "...into Equation 5 yields..."

Line 192 - figures: Figures 1 and 2 are introduced here, but figure 2 is not actually discussed until after discussion of figure 4 breaking the flow of discussion and figures, and leaving me, initially, confused as to how I'd missed the discussion of figure 2. In fact, only the general environmental conditions shown in figure 1 are discussed here, not the gas flux results, which are discussed much later. Figure 1 would be better split into 2, separating the gas fluxes into a figure matching the format of current figure 2. The figures showing the gas flux results could then be placed in a logical order within the discussion. Since wave state is a relevant parameter in the later discussion, it would be useful to add a time series of at least significant wave height to figure 1.

Line 207: The authors note that their estimates of whitecap fraction as a function of wind speed are substantially lower than other recently published values – at times an order of magnitude lower. A likely reason for this is the exposure settings on the camera. During the HiWinGS project cruise in 2013 two independent sets of cameras were used for whitecap imaging. They were initially found to give whitecap fractions that differed by a factor of several. Tests were conducted during the final transit of the cruise, in which a pair of identical cameras were run side by side; one with fixed exposure settings, the other having the exposure settings changed every few hours. The exposure settings were found to make a substantial difference to the whitecap fraction calculated using the same Callaghan and White (2009) algorithm used here – up to a factor of 4 for the range of settings tested. It was found that almost all of this difference (both between the 2 cameras in the exposure trial, and between the two sets of cameras used throughout the cruise) was removed if the images were first 'normalised' to remove any brightness gradient across the image. Brief details of these tests will be given in

Brumer, S. E., C. J. Zappa, **I. M. Brooks**, H. Tamura, S. M. Brown, B. Blomquist, C. W. Fairall, A. Cifuentes-Lorenzen, 2017: Whitecap coverage dependence on wind and wave statistics as observed during SO GasEx and HiWinGS, *J. Phys. Oceanogr*. (under revision)

Line 213-215: "Stage A whitecap fraction data is highly variable at ~11 m s-1 213 wind speeds (Figure 3b), which is driven by the difference in the wind-wave conditions during Knorr_11 (ST184 vs ST191, Figure 4a)" – two points:
(1) the difference is ascribed to different wind-wave conditions at the two stations, but no wave data are shown. As noted above, relevant wave parameters need to be added to figure 1.
(2) A similar broad range of stage A whitecaps is evident at around U = 6 m s^{-1}, also resulting from grouping of high/low values by different stations...are the wind-wave conditions similarly different in this case?

Lines 218-223: The authors first note that where stage A whitecap fractions is < 10^{-4} the relationship with R_H is more scattered than at higher fractions; they then note a number of factors that affect wave breaking and so whitecap fraction, but don't make a coherent link back to their initial point about the scatter in the stage-A whitecap / R_H relationship. This reads as an almost unconnected series of statements...all true, but leaving the reader wondering what the point being made is.

Line 282: "...(Figure 5)" -> "...(Figure 5b,d)" – again text refers to high wave conditions for ST19 but no wave data provided for reader to assess.

Figure 5 and the discussion of it have some general issues:
- It's hard to see the pink/green lines against the mass of pink/green dots on panels a and b – it would help here to plot the dots in a paler shade of pink/green to allow the lines to stand out.
- The curves shown, for the COAREG3.1 model are a useful reference, but fits to the actual data are also needed; these would allow a much clearer assessment of how closely the COARE model agrees with the observations.
- lines 282-284: "Under the high wind, high wave conditions encountered during ST191, the wind speed-dependence of $k_{DMS,Sc}$ was lower than expected, with a slope roughly half that of the rest of the cruise data. This effect was not observed at ST184." – since the ST191 data are not highlighted in any way it is not possible for the reader to judge the behaviour here. Of note perhaps is not simply the high wind and wave conditions during ST191 but the different time history of the winds – a sustained period of high winds during ST191 vs a very short period in ST184 where the wind rises rapidly, spikes, and decreases rapidly. These two periods are likely to produce very different wave fields at the same wind speeds – again, reason to plot the wave parameters in figure 1 – which might explain the very different whitecap fractions seen in Figure 1b for these periods.

Line 294 & 305: the phrasing "until 11 m s^{-1} wind speed" is rather clumsy; 'until' implies a variation over time, which is not what is meant – "...up to wind speeds of 11 m s^{-1}" would read better.

Line 326: "...Δkw is near zero at very low wind speeds (U10 ≤ 4.5 m s^{-1})..." – this is hard to judge. Eyeballing the data points I would agree; however, there are only 3 points at U < 4.5 m s^{-1}, and all of those at U>3 m s^{-1}. Their mean Δk is ~5 cm/hr and the fitted curve approaches a Δk of ~3 cm/hr at U = 0, not zero. One might argue for an alternative functional form in which the exponents were not prescribed might better represent the data; the quadratic used here implicitly assumes the functional dependence. (Note also, the figure caption states that the plotted curve is cubic not quadratic).

What is the reasoning behind using 4-hour averages of transfer velocities here (and elsewhere)? 4 hours is quite a long time relative to the time period over which significant changes in forcing can take place. Granted it greatly reduces scatter, but I would wary of averaging over periods much long than ~1 hour.

Line 358: "...the relationship between Δkw and whitecap areal extent appears to be linear." – I'm not convinced this is entirely true – approximately so over the range 0.005 < W < 0.05, but ~half the data points lie at W < 0.005, and seem to drop off rather more rapidly than the fit to the high values of W would indicate. Plotting W on a log scale might give a rather different impression. Not that IF the relationship is linear as suggested then eyeballing a fit (if you claim a linear fit, it would help to show it!) suggests Δk = 10-15 cm/hr at W = 0, which raises questions as to why that should be when there are no bubbles to account for a difference in k, and why this minimum difference is several times higher than that derived as a function of wind speed. If on the other hand a roughly linear fit of Δk to log(W) existed (which I think the rapid drop off in Δk at very low W might support) then Δk would approach zero at low W.

Line 366-367: "In this case, Δkw should be more strongly correlated with *WA* than *WB* or *WT*." – in a general sense, this is true, but is only if the various factors affecting foam decay vary. If foam decay rate is constant then W_T should be proportional to W_A.

Line 387: "Both models significantly underestimate $k_{bub,CO2}$ at wind speeds below about 11 m s^{-1}." – Actually both models rather underestimate the observationally derived $K_{bub,CO2}$ at all windspeeds; however, note that both models are driven by the observed wind-whitecap relationship, which has already been stated to be low compared to other recent estimates. Is the agreement better using a whitecap function that agrees more closely with the recent consensus?
While the Asher et al. model is lower than the observations, it is not wildly so (essentially matching the lower boundary of the observed values), and the agreement in both values and functional behaviour is rather convincing.

Line 455: "...eddy covariance setup..." -> "...eddy covariance system..."

Line 463: reference to paper in preparation not generally allowed...if it's 'in press' by the time this manuscript it copy edited, the copy editor will want the reference cut.

Figure 3: it's hard to pick out the curves and black square points against the mass of black dots. Suggest changing black dots to mid-grey and ensuring everything else is plotted over them. It would also be good to see a functional fit to this data as well as the functions from previous studies...especially as this function is later used to drive the $k_{bub,co2}$ models plotted in figure 8.