

We thank both reviewers for their positive and constructive comments. We address their specific points below:

### **Reviewer 1: Bill Asher**

*I think the authors are glossing over a potential problem in that in the system they are studying, CO<sub>2</sub> is an invasive flux (air-to-ocean) and DMS is an evasive flux (ocean-to-air). My hunch is that Equation 5 is only strictly true when both gases are far from equilibrium \*and\* the flux is in the same direction. Problems arise in applying Equation 5 for a mixed system, where one gas is invading and one is evading, because the bubble gas flux is not the same, even when normalized to a common diffusivity/solubility. In the case of invasion, the bubble overpressure drives more gas than expected (based on the bulk air-ocean concentration difference) into the water.*

This is true, and we are glad that you brought it up. However, it is a small effect. Woolf (1997) provides the means to calculate the magnitude of this effect. The average fractional extra pressure on the gas in contact with the sea ( $\Delta$ ) can be estimated from:

$$\Delta = (U/U_i)^2\%$$

where  $U_i$  is the wind speed at which the supersaturation of a particular gas equals 1% (49 m s<sup>-1</sup> for CO<sub>2</sub>). A high wind speed (20 m s<sup>-1</sup>) gives  $\Delta = 0.167\%$ . An atmospheric pCO<sub>2</sub> of 400 ppm implies that the bubble overpressure would be 0.67 ppm. This is a ~2% enhancement of the CO<sub>2</sub> flux when the air/sea concentration gradient is small (minimum for this study = 30 ppm). Larger air/sea concentration gradients would diminish the magnitude of the bubble overpressure further.

We have added the following text to our revised manuscript after equation 5:

“Strictly speaking, Equation 5 should also account for the influence of bubble overpressure, which alters the gas flux due to bubbles when the concentration gradient is toward the ocean. The extra pressure on the gas in the bubbles is calculated following Woolf (1997):  $\Delta = (U_{10}/U_i)^2 \%$  where  $U_i$  is the wind speed at which the supersaturation of a particular gas equals 1% (49 m s<sup>-1</sup> in the case of CO<sub>2</sub>). A high wind speed (20 m s<sup>-1</sup>) gives  $\Delta = 0.167\%$ , which would lead to a ~2% enhancement of the CO<sub>2</sub> flux when the air/sea concentration gradient is 30 ppm (minimum for this study) and into the ocean. The magnitude of this effect would be larger for gases less soluble than CO<sub>2</sub> but we are able to ignore it for the purposes of this study.”

*one thing is clear from looking at the material in the supplements, is that using the Asher et al. (2002) relationship for both CO<sub>2</sub> (invasion) and DMS (evasion) is not correct. The Asher et al. (2002) relation is only for invasion. For evasion, there is a separate equation in Asher and Wanninkhof (1998).*

This is also true and we are glad that it has been pointed out. As the bubble term for DMS is small, there will be negligible impact upon our data. To be absolutely correct, we have adjusted our data and the relevant equations in the revised manuscript.

*Line 54: "These processes include ..." Comment: Buoyancy effects are not a process. It might be better in this sentence to say something like "These processes include diffusion, surface renewal, and bubble-mediated transport. In turn, turbulence can be generated by wind stress, wave-induced mixing, buoyancy currents, and wave breaking."*

Change made.

*Line 56: "A variety of theoretical, laboratory, and field ..." Comment: I don't think this sentence is strictly true. My opinion is we have a fairly good understanding of the factors that affect gas exchange from a phenomenological standpoint (the authors list them just a couple of sentences earlier). What we lack is how to determine which of those processes are important under a given set of circumstances.*

Changed sentence to:

“A variety of theoretical, laboratory, and field approaches have been used to study the processes that control air/sea transfer, but we do not yet have a firm understanding of their relative importance under a range of atmospheric and oceanic conditions.”

Line 60: "Gas transfer via bubbles ( $k_{bub}$ ) ..." Comment: It would be good to define  $k_{bub}$  here. The point is that there are a couple of different ways to do this, you can go the Memery and Merlivat (Memery, L. and L. Merlivat (1985). "Modeling of the gas flux through bubbles at the air-water interface." *Tellus, Ser. B* 37: 272-285) approach and use the bulk air-water concentration difference and accept that  $k_{bub}$  for invasion and evasion are different (e.g., I used this approach in Asher et al. (1996, *JGR-Oceans*)) or you can redefine the air-water concentration difference in terms of how bubbles would affect the equilibrium and have a common  $k_{bub}$  (but then it might get complicated relating  $k_{bub}$  for invasion and evasion) as done by Woolf (1997).

We have clarified the definition of  $k_{bub}$  here and cover the issue of invasion vs evasion in detail on Line 161 (to address the comment below).

Line 78: "These measurements typically show DMS gas transfer velocities that are lower and exhibit more linear wind speed dependence than those estimated for CO<sub>2</sub> based on dual tracer studies (e.g. Bell et al., 2015; Yang et al., 2011; Goddijn-Murphy et al., 2012)." Comment: I think the authors should be clear here that there are no CO<sub>2</sub> measurements from dual-tracer studies. There are DT measurements for SF<sub>6</sub>/He, which get related to CO<sub>2</sub> through diffusivity. Then there are EC measurements for CO<sub>2</sub>. Comparison of the DT-derived CO<sub>2</sub> transfer velocities with CO<sub>2</sub> transfer velocities produced by EC measurements of CO<sub>2</sub> fluxes shows relatively good agreement. It is the transfer velocities produced by EC measurements of DMS fluxes that show different behavior.

Change made.

Line 87: Comment: maybe want to note that they agree when normalized to a common diffusivity.

Change made.

Line 126: "The air side gas transfer contributes about 5% on average to the total resistance for DMS."

Comment: The air-side resistance fraction is a function of wind speed. Does this 5% increase as  $U$  increases? COAREG must reproduce this, it was measured by McGillis et al. a while back.

McGillis et al.. (2000) used a non-linear relationship between waterside transfer velocity and wind speed. Based on more recent measurements of DMS gas transfer velocity it is more appropriate to assume a linear relationship. As a result, the relative contribution of airside resistance to total resistance does not change substantially as a function of wind speed (see Supplemental material in Bell et al 2015).

Line 161: the relation in the text showing  $k_w = k_{int} + k_{bub}$ . Comment: I wonder if maybe it is time to stop writing this as a general expression (I know, I am guilty of this as well). What is generally true is that the total gas flux is equal to the sum of interfacial flux and the bubble flux. Saying the overall transfer velocity is equal to the sum of the two transfer velocities really only works if the concentration difference is far from equilibrium. Work through David's relations from the 1997 paper and you'll find they are a bit convoluted in terms of how exactly the pieced (his Delta term) fit together to make a coherent physical picture. If you start by assuming it is the fluxes, not the transfer velocities, which sum linearly, the assumptions required to get to the various relations proposed are more easily understood.

We have changed the text to be more accurate.

Technical Comments:

1. Multiple citations are not in any recognizable order. Sometimes they are chronological, sometimes alphabetical. I don't remember what the ACP style guide says, but I am sure it is not "random." Agreed! We will correct this.

2. Line 74: "... studies indicate a non-linear dependence ..."

Changed to "studies observed a non-linear wind speed dependence"

Line 91: Shouldn't cite papers that are not published or submitted.

OK, removed.

*Line 175: The two f values are opposite from what is given in the supplement. Not sure which is correct, but it should be consistent (and correct).*  
Good spot. Changed in the supplemental material.

*Line 323: I know this is petty, but I don't think Woolf (1997) is based on laboratory data.*  
Woolf (1997) did use laboratory data from other studies. We will clarify this.

*Equation 7: Figure caption says "cubic" and equation 7 is quadratic. Resolve this difference.*  
Changed.

*Line 384: The citation to Asher and Wanninkhof (1998) should be to Asher et al. (1996). If you really must cite Asher and Wanninkhof (1998) in this context, which you shouldn't, at least make it the other Asher and Wanninkhof (1998) paper that is directly relevant (see citation above).*  
Changed.

## **Reviewer 2: Ian Brooks**

*Line 86-87: "In that study, no statistically significant difference was observed in gas transfer-wind speed relationships of CO<sub>2</sub> and DMS for winds below 10 m s<sup>-1</sup>" – need to clarify this statement, was a significant difference found for winds above 10 m s<sup>-1</sup>? Was 10 m s<sup>-1</sup> the maximum wind speed in the study?*  
Changed to:

"In that study, no data were collected for winds greater than 10 m s<sup>-1</sup> and no statistically significant difference was observed in the CO<sub>2</sub> and DMS gas transfer-wind speed relationships."

*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).*  
This has been corrected.

*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<sub>a</sub>. Any uncertainty in this will impact the later results and should be acknowledged and if possible quantified.*  
We have added the following sentence:  
"Note that the use of the COAREG 3.1 model introduces a small uncertainty in our estimates of waterside DMS gas transfer velocity (approximately  $\pm 2\%$  when wind speed = 20 m s<sup>-1</sup>)."

*Line 126-127: "The air side gas transfer contributes about 5% on average to the total resistance for DMS" – do you mean 'air-side resistance' rather than air-side transfer?*  
Yes. Changed.

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

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

*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.*

We agree. Figure 1 has been split and a timeseries of Hs has been added. Figures are reordered so the gas flux/transfer velocity figures follow the whitecapping figures.

*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)*

Many of the potential issues highlighted above (exposure settings, intercomparability of image processing) have actually been addressed. We have added additional information about the whitecap image processing to the Supplemental material.

The lower bounds of the Knorr  $W$  data do fall below recent parameterisations, most notably in the wind speed range of  $\sim 7.5 \text{ m s}^{-1}$  to  $12.5 \text{ m s}^{-1}$ . However, when the Knorr  $W$  data are binned by wind speed (squares in Figure 3a, main manuscript), the binned  $W$  data compare favourably with the Schwendeman and Thomson (2015) estimate of whitecap fraction at all wind speeds, with the exception of the lowest bin of  $3 \text{ m s}^{-1}$ . The binned Knorr  $W$  data also agree favourably with Callaghan et al., (2008), except for the  $9 \text{ m s}^{-1}$  and  $11 \text{ m s}^{-1}$  binned datapoints, for which the Knorr binned  $W$  data are lower. When binned by wind speed, the Knorr  $W$  data and the two parameterisations generally fall within a factor of 2 across the wind speeds examined.

There is clearly quite a lot of scatter in the Knorr  $W$  dataset, and many data points lie below the Schwendeman and Thomson (2015) and Callaghan et al., (2008) parameterisations. However, we do not believe that the primary driver of the differences observed is due to the image processing methodology employed. Rather, it could be that other sources of variability (at a given wind speed) have caused the observed differences. Examples include: (i) water chemistry (surfactants); (ii) total wave field energy dissipation; and (iii) energy dissipation by microscale breaking waves as opposed to air-entraining whitecaps. It is beyond the scope of this paper to address these potential causes.

*Line 213-215: “Stage A whitecap fraction data is highly variable at  $\sim 11 \text{ m s}^{-1}$  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.*

Done.

*(2) A similar broad range of stage A whitecaps is evident at around  $U = 6 \text{ m s}^{-1}$ , also resulting from grouping of high/low values by different stations...are the wind-wave conditions similarly different in this case?*

We agree that there is a grouping at  $U=6 \text{ m s}^{-1}$ . It is unclear what caused this (although ST181 was in the Gulf Stream and thus had much higher water temperatures). However, this discussion is beyond the scope of this paper.

*Lines 218-223: The authors first note that where stage A whitecap fractions is  $< 10^{-4}$  the relationship with RH 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 / RH relationship. This reads as an almost unconnected series of statements...all true, but leaving the reader wondering what the point being made is.*

We agree that this text needed adjusting. We also decided that the text discussing Stage A whitecap variability and  $R_H$  was not essential to the manuscript and have moved the modified text and the related Figure to Supplemental information.

*Line 282: "... (Figure 5)" -> "... (Figure 5b,d)" – again text refers to high wave conditions for ST191 but no wave data provided for reader to assess.*

In this instance, we feel that it is sufficient for us to reference the in depth discussion in Bell et al. (2013).

*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 figure has been adjusted to improve readability.

*- 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.*

We tried this but it made the plots too busy. We decided it best not to include both.

*- 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.*

In addition to plotting the wave data, we have added the word 'sustained' to this sentence and referenced Bell et al. 2013)

*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.*

Change made.

*Line 326: "... $\Delta k_w$  is near zero at very low wind speeds ( $U_{10} \leq 4.5$  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  $\Delta k$  is  $\sim 5$  cm/hr and the fitted curve approaches a  $\Delta k_w$  of  $\sim 3$  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.*

We have tested a variety of functional forms and number of exponents for our best fit: linear, polynomial (n=2,3), power (n=1,2) and exponential (n=1,2). The goodness of fit was extremely comparable between all fits (from  $R^2 = 0.66$  to  $R^2 = 0.67$ ) with the exception of the linear fit ( $R^2 = 0.62$ ). As the goodness of fit did not help with our decision, the choice of fit is subjective. Having considered the different fit lines, we feel that a simple power law fit ( $\Delta k_w = 0.177U_{10}^{1.928}$ ) represents the low  $\Delta k_w$  data better than our original choice of fit. We have changed the manuscript accordingly.

The fit line to the data tells us that  $\Delta k_w$  is indeed positive at low wind speeds. This could be driven by effects such as chemical enhancement and sea surface skin temperature (see Discussion), which we have not controlled for here. We used the term 'near zero' intentionally but now clarify this in the revised manuscript with "( $< 4.5$  cm/hr)"

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. Some averaging of the data was necessary to reduce the scatter, but we accept that minimising the averaging time period is important. We have changed our averaging period to two hours.

Line 358: “...the relationship between  $\Delta k_w$  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  $\Delta k = 10\text{-}15$  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  $\Delta k$  to  $\log(W)$  existed (which I think the rapid drop off in  $\Delta k$  at very low  $W$  might support) then  $\Delta k$  would approach zero at low  $W$ .

We have revised the statement to:

“The functional form of the relationship between  $\Delta k_w$  and whitecap areal extent appears to be linear for  $W_T > 0.005$ . However, the Knorr\_11 dataset is small and quite scattered, particularly when  $W_T < 0.005$ .”

Line 366-367: “In this case,  $\Delta k_w$  should be more strongly correlated with  $W_A$  than  $W_B$  or  $W_T$ .” – 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  $W_A$ .

We agree. We feel this case is broadly covered later in the paragraph. We have adjusted the text slightly to take into account a constant decay rate:

“ $W_T$  and  $W_A$  may be equally good (or poor) proxies for bubbles because: (i) surfactant activity was either insignificant or sufficiently invariant in the study region (despite high biological productivity) that  $W_B$  does not confound the relationship between  $W_T$  and  $W_A$ ”

Line 387: “Both models significantly underestimate  $k_{bub,CO2}$  at wind speeds below about 11 m s<sup>-1</sup>.” 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?

The agreement is slightly improved (at intermediate wind speeds) using a different whitecap function. Although we see no reason to doubt our whitecap measurements, it is useful to observe the importance of the wind speed-whitecap fraction relationship to the output from these models, so we have added these  $k_{bub,CO2}$  estimates to the manuscript.

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.

We are possibly less optimistic on this point and hopefully can agree to disagree!

Line 455: “...eddy covariance setup...” -> “...eddy covariance system...”

Change made.

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

Change made.

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 kbub,co2 models plotted in figure 8.*  
Revised accordingly.