Author's reply to 'Interactive comment on "Air-sea gas exchange at hurricane wind speeds" by Kerstin E. Krall and Bernd Jähne' by Christopher Fairall

September 3, 2019

The authors thank Christopher Fairall for his thorough review and helpful comments. A point by point answer to his questions and comments can be found below.

Missing bubble contribution

> Reviewer's comment: Their main finding – that bubble transfer is negligible compared to the direct transfer for all gases up to wind speeds of 30 m/s – is surprising (at least to this reviewer).

This was very unexpected for the authors as well. This is why we reported the Kyoto results only on conferences but did not write a paper and waited for the results of a second experiment in the much larger SUSTAIN facility with real seawater, where very similar results were obtained.

Reconciliation of lab and field measurements

> Reviewer's comment: The authors' conclusions are difficult to reconcile with open ocean measurements but I leave it to them to consider how to handle that. In my opinion they are too dismissive of the large number of experimental (tracers and eddy covariance) studies that indicate insoluble gases and CO2 have substantially higher transfer velocity above 15 m/s. I question that the Zavarsky paper is sufficient cover.

In the conclusions of our paper, we will put more emphasis on the unsolved problem of wave age or fetch dependency of the gas transfer velocity.
By the way, the authors are surprised that both reviewers do not comment at all our findings for wind speeds beyond 33 m/s, were we found a very steep increase of the gas transfer velocities even without the effect of bubbles, being associated with various rapid surface fragmentation processes at the free surface. We do not claim that this effect happens in the very same way at the open ocean, but it will happen also there, indicating that also the transfer of all water-side controlled gases will be enhanced significantly independently of solubility. In our view, this is an important new finding with relevance for the global fluxes between ocean and atmosphere.

Computation of u_{10} and momentum balance

> Reviewer's comment: I don't understand why they used Powell's open ocean estimates of Cd when they could have used Donelan's 2004 results actually determined in the Miami wind tunnel. Also, please give us a sentence explaining Takagadi's method for getting friction velocity so we don't have to go look it up. I am guessing they assumed a momentum balance at the interface to compute u*w from u*a (square root of ratio of air to water density). This assumes that the growth of the wave field has negligible effect of the momentum balance. Is this right?

Also, they switch back and forth between u*w and just u* - I assume they mean the same thing. Just be advised that it is quite a stretch from a setting on an instrument dial to actual waterside friction velocity.
and later ...
U*a is a measure for momentum extracted from the wind. Some of that momentum is realized in the ocean via direct viscous transfer at the interface, some goes into growing waves and some is later realized as turbulence when waves break. If locally waves are in dynamic balance, then the momentum flux from the air is the same as the momentum flux realized in the ocean. So, how close is the balance in a wind tunnel?

- We now explain this in more detail in the manuscript. To this end we will add our co-workers from Kyoto (Naohisa Takagaki) and Miami (Andrew Smith) as co-authors, because they made the measurements and greatly helped with computing u_{10}. We decided to use Donelan et al. (2004) (lab measurements of C_D) instead of Powell et al. (2003) (field measurements of C_D) to convert measured wind speeds in Miami to $u_{*,a}$ and u_{10} and we also found a mistake in converting between $u_{*,a}$ and $u_{*,w}$, which we corrected for the final paper. This slightly changes the relationships $k_x(u_{*,w})$ and the model parameterization equations, but not our findings.

- In short fetch wind-wave tunnels, the growth of waves has a considerable effect on the momentum balance. At the moment, we have only data from one fetch, measured in the large Marseille facility and only being published in a PhD thesis (Bopp, 2018) with results similar to Banner and Peirson (1998). So the best we could do is to assume momentum balance. We wanted to use the water side friction velocity, because gas transfer is controlled there.

- Indeed, the label in Fig. 5 should read as u_{*w} and not u_* and will be changed in the final paper.

- As a side note, the wind-wave tank that Donelan et al. (2004) used was a different one than the one used in this study (SUSTAIN). So Donelan et al. (2004) did not determine C_d in **the** Miami wind wave tunnel, but merely in **a** Miami wind wave tunnel.

- In the fetch-limited SUSTAIN tank, waves are growing until their structure changes/collapses due to momentum transfer (forcing) from the wind or breaking and they do not reach true equilibrium where air-side and water-side stress are exactly equal. An upcoming experiment in SUSTAIN on the drag coefficient and momentum balance (lab vs. field) is presently in planning stages.

DMS and carbon dioxide

Reviewer's comment: Another example, McNeil and D'Asaro (2007) did not 'measure' gas transfer velocities, they inferred them from water concentration measurements. Furthermore, their basic assumption in the analysis is that both free surface and bubble transfer velocities scale with u*w and nothing else. However, there is considerable evidence that the air volume flux from breaking scales as u* and other wave parameters (see Deike et al. 2017). Also note that Deike and Melville (2018) used this approach to estimate kb for DMS and CO2 – treating both gases as highly soluble. They present measurements of bubble area from the wind tunnels but no estimates of volume flux. I think the bubble volume flux data should appear in Fig. 7c. Finally, the authors might note that Rhee et al. 2007 found considerable enhancement of k for insoluble gases when bubbles were introduced.
and later ...
The discussion of field measurements of k for DMS and CO2 is very useful (Section 4.5). It also illustrates the rather inconclusive state of field observations. The data from Zavarsky et al (2018) show essentially no difference between CO2 and DMS. The analysis of Fairall et al. (2011) which compiled all the direct flux data to date showed significant differences. The HIWINGS data shown in this figure are quite surprising for CO2. Blomquist gives k CO2 a power dependence of u10^1.68, which is not linear. Because of the conditions, I don't think the HIWINGS data below U10 of 10 m/s should be considered. Even ancient information such

as Wanninkhoff's famous formula indicate a quadratic wind speed dependence for insoluble gases. Earlier suggestions that k CO2 should scale as U^3 were based on the assumption that whitecap fraction scaled as U^3. More recent observations have shown that this is not the case, with much weaker wind speed dependence at high wind speeds. I think the authors may be placing too much importance on Zavarsky. From Wanninkhoff's radioactive tracers, to a number of deliberate tracer studies, and perhaps all other eddy flux measurements CO2 goes at least quadratic with wind speed. At u10=18 m/s, the value is close to 100 cm/h for open ocean measurements.

- For the data of McNeil and D'Asaro (2007), we will change 'measured' to 'estimated'.
- We are surprised about the statement that the bubble volume flux V_a should scale linearly with u_*. Did you really mean this? The equations in Fig. 4 in Deike et al. (2017) rather says $V_a = 2.3 \cdot 10^{-3} c_p^{-0.9} u_*^{1.9}$. By the way, this finding is just the opposite of the finding by Blomquist et al. (2017). They found lower gas transfer velocities in 'young' sea states than in 'old' seas for carbon dioxide and attributed this to higher bubble contributions at older seas. Deike et al. (2017) found that the air entrainment is much lower at high wave ages. The effect is large, air entrainment scales roughly with the inverse wave age. Thus the wave age dependency is still an open question and further investigations are required.
- Rhee et al. (2007) is, among other studies, rather irrelevant for our work, because 1) their highest measured wind speed is 13 m/s, and 2) their means of bubble generation (submerged aerators) is very different from ours (wave breaking induced bubbles only). Therefore such a comparison is not meaningful. In the introduction, we refer, of course, to the two previous lab studies in the Kyoto high wind speed facility: Iwano et al. (2013, 2014) and Krall and Jähne (2014).
- We selected the data of Blomquist et al. (2017) because it had the highest wind speeds up to 25 m/s and those of Zavarsky et al. (2018) because they show no difference is the gas exchange rates between DMS and carbon dioxide. We place equal importance to both field studies, and can only state that there are contradicting results. Unfortunately, eddy covariance measurements are still not precise enough (especially compared to gas transfer velocity measurements in the lab) and prone to systematic errors. Our current conclusion is therefore, that there are significant yet unresolved wave age and sea state effects. We will emphasize this more clearly in the conclusions and the abstract of our paper.
- We will add a short paragraph to the manuscript, discussing transfer velocities measured using the dual tracer technique with He and SF_6, which have a very low solubility and should, therefore, have a larger bubble effect than CO_2. However, SF_6 and He were found to have lower transfer velocities than CO_2 measured with eddy covariance, see the compilation of field measurements in Garbe et al. (2014, Fig. 2.10a). This is another contradicting field result.

Minor comments

Reviewer's comment: Bubbles may also suppress turbulence through density stratification.

Correct, but for the wind-wave tanks with just below one meter depth, this effect is not significant at all for our measurements.

Reviewer's comment: Eq (5) This terminology is confusing with the un-numbered equation on Page 2 line 14.

We changed the wording in the first paragraph of Sec. 2.1 for clarity.

Reviewer's comment: Page 4 line 1 Suggest referencing bubble model work of Liang et al. GLOBAL BIO- GEOCHEMICAL CYCLES, VOL. 27, 894–905, doi:10.1002/gbc.20080, 2013 and ear- lier work.

We decided not to cite Liang et al. (2013), because their focus is more on supersaturation and fluxes and they did not measure the transfer velocity.

> Reviewer's comment: Page 5 Eq (7) I am confused by the terminology. Can Qb be volume flux and Qb/As also be a volume flux? If we equate kr with the volume of air ingested per unit area per unit time (units velocity), then that should be on the order of 30 cm/hr at u10=15 m/s and u*w=2 cm/s (see, Deike et al, 2017). That does not compare well with Fig. 7 c, where kr doesn't reach those values until u*w is greater than 10.

- Q_b/A_s has the units of a velocity. We will change its name in accordance with Deike et al. (2017) to 'air entrainment velocity'.
- Deike et al. (2017) found a considerable wave age effect and our measurements add another value for very low wave ages not covered by the data Deike is using.

> Reviewer's comment: Page 5 Eq (10) This equation is similar to Woolf parameterization. For the volume flux is kr=2450*whitecap fraction (cm/h) and the parameter e=kc/kr*sqrt(600). How does this compare to your results?

We can't directly compare because we have not yet estimated the whitecap fraction. We will do this in a further paper once we have analyzed the images from the water surface. Currently, we can only say that the solubility dependency is about the same. However, Mischler (2014) has shown in a bubble tank study, that the Woolf et al. (2007) and the Mischler (2014) parameterizations for the bubble mediated transfer velocity k_c perform equally well, however, the Mischler (2014) parameterization uses one fewer parameter.

> Reviewer's comment: Page 7 section 2.3. This discussion of droplet effects is a little confusing. It seems to me that ejecting a droplet does not change the waterside concentration, so their measurement method does not capture it. If the drop has time, it would transfer gas to the air and that would reduce free surface transfer further down the line. Is that what they are trying to say? This argument about time scales ignores the fact that the droplets leave the wind tunnel before they can do much transferring – this is discussed in Andreas and Mahrt 2016.

Even though the fetch of the wind-wave tanks is rather short compared to open ocean conditions, some of the droplets generated, will impact the water surface again. Andreas et al. (2017) discusses the time scales involved in great detail. They state, that only for the largest radii and for weak winds less than 15 m/s, the droplets fall back into the ocean before they establish equilibrium with the atmospheric gas reservoir. So even though not all spray droplets reimpact the water surface since they are blown out of the wind wave tank, those that do impact the water again do change the water side concentration, since they will have equilibrated with the air.
Since, according to Andreas et al. (2017), at extreme conditions, the spray droplets do come into equilibrium before falling back into the water, the diffusion coefficient of the tracer no longer plays a role in spray mediated gas transfer. Therefore, Helium, despite having a much higher diffusion coefficient than other gases, will no longer have a correspondingly faster transfer velocity across the pray droplet interface, since this transfer velocity only depends on how much spray is generated.

> Reviewer's comment: Figure 10. What drag coefficient is used for the curve shown for modeled DMS and CO2?

We used the u_{10}-$u_{*,w}$ relationship shown as a gray line in Fig. 4a, which corresponds to the drag coefficient shown as the gray line in Fig. 1 below.

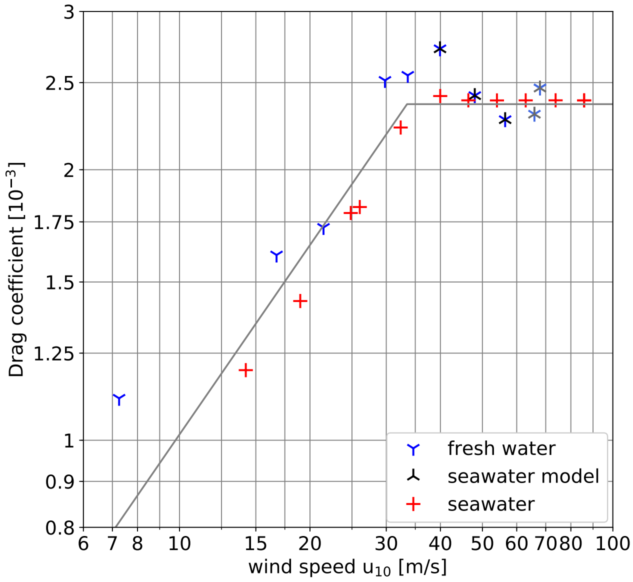

Figure 1: The gray line shows the drag coefficient used for converting $u_{*,w}$ to u_{10} for the Figs. 9, 10 and 11 in the manuscript.

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