Interactive comment on “Measuring air–sea gas exchange velocities in a large scale annular wind-wave tank”

The authors would like to thank the anonymous referee 2 for reviewing our manuscript and for the valuable comments and suggestions which have improved the quality of this publication. Our point by point response to the comments, is listed below.

General comments:

- *This paper is fairly well written and the approach is well explained. The results are interesting and should be published. There is obviously more to come from the experiments than is presented here. Whilst I am comfortable with the authors introducing this paper as the first of many, I do not see why they feel the need to discuss the other 12 chemical species if only data from two are presented? The authors' subsequent papers that discuss these other 12 compounds will easily be able to reference the calculations and experimental methodology presented in this paper. I therefore recommend removal of a lot of the references made to the other 12 compounds.*

Answer

The large number of gases which were measured in parallel is innovative and characterizes the proposed method.
"For the first time, parallel measurements of total air and water-side transfer velocities for 14 individual gases within a wide range of solubility, have been achieved."
No previous study has investigated the transfer velocity of so many compounds in parallel or has focused on the middle and low solubility range using such a variety of tracers. We focus on two contrasting tracers (the sparingly soluble N_2O and the highly soluble CH_3OH) to illustrate the performance of our method. The distinguishing aspect of this study (and methodology) though, is the majority of tracers being measured in parallel, the great piece of information such measurements embody, the step forward in understanding the mechanisms behind the gas exchange. The authors believe that the present publication, as the first of two by this first author, should include the complete information, in particular as the follow up paper uses all the data. This makes it easier for following publications to focus more on the physical aspects of the gas exchange avoiding many introductory details.

- *Within the paper, my main concern is the presentation of the low surfactant data. It is very interesting that the surfactant effect can be observed in the water-side controlled gas measurements and cannot be seen in the air-side controlled gas data. However, without direct u* measurements, the inferences that might be made from Fig 9 are driven by an (assumed) interpolation between clean and high*

surfactant u data. This suggests a trend that was not in fact measured. I recommend that these data are either removed or presented in a way that does not rely upon the u* data (and thus an interpolation).*

Answer The authors considered this point carefully and decided to present the transfer velocity data of Fig. 9 against the relative wind speed, u_{ref}. The reason for this, is that u_{ref} is the only stable parameter, common in all seven experiments (i.e u_* and mss change under surfactant covered surfaces). Additionally, a linear scale of each plot is introduced in Fig. 9 (as requested in the last specific comment).

Regarding the missing (and therefore interpolated) u* measurements of the low surfactant concentration, a solution is provided. After the evaluation of the latest dataset (Bopp 2014), the u_* data of exactly the same experimental conditions with the missing low surfactant case are now available and replaced the old values. (i.e all plots tables and related text are updated using the latest and more accurate u* values).

- *Further, the manuscript needs some discussion of breaking waves and bubble formation in the Aeolotron tank. At what wind speed or u* value does/did this occur? How well/poorly does it reflect the real world? This is highly relevant when the authors begin to compare their data with other gas exchange parameterizations that have been derived from field experiments.*

Answer
These phenomena are discussed in paragraph 4 of section 3.2.5.

Specific comments:

- *Page 1644, Lines 14-15: Change 'In contrast, the surfactant affected CH3OH, the high solubility tracer only weakly.' to 'In contrast, the surfactant only weakly affected the high solubility tracer (CH3OH).'*

Answer: Text has been corrected accordingly.

- *Page 1644, Line 14: Suggest adding 'the relatively insoluble' before N2O*.

Answer: Text has been introduced.

- *Page 1645, Lines 4-8: This description is fine, but I think an equation showing the relationship between the different terms would be useful here.*

Answer: The authors would prefer to leave it this way. We believe that the addition of an equation would complicate the sentence here.

- *Page 1646, Line 4: I would like to see some discussion of the eddy covariance technique here. The discussion of the measurement frequency limitations of the dual tracer approach is useful, but eddy covariance is also capable of resolving gas transfer estimates on short timescales. For example, the recent work by Yang et al (referenced later in the text) has provided novel estimates of soluble gas (methanol) transfer velocities. Other recent work by Bell et al., (ACP, 2013) suggests a possible role for wind-wave interactions in determining gas exchange. Further, the utility of using multiple gases to understand physiochemical controls on gas exchange is not restricted just to wind-wave tanks. Eddy covariance offers this opportunity too (e.g. Miller et al., GRL,2009).*

Answer: In contrast to referee 2, here referee 1 suggested less discussion related to other transfer velocity measurement techniques.
Paragraph 4 discusses previous studies which use a mass balance approach and does not focus on the eddy covariance method. In paragraph 3 of the introduction, we generally refer to other transfer velocity studies separating them to lab and field studies. The reader is referred to previous reviews for more details. As our method is based on a mass balance approach, in paragraph 4 we refer specifically to mass balance methods commenting on two general drawbacks: the time resolution and the focus on the sparingly soluble tracers. Here as well, we are not going into further detail.

- *Page 1646, Line 17: Change 'show clearly different' to 'clearly show different'*

Answer: Changed

- *Page 1647, Lines 12-18: Please define the various terms within the text immediately preceding eqns 1-4. Having them in the legend of Figure 1 is useful but I would prefer that the information also be repeated here.*

Answer: All additional information presented in the legend is now also given in the text.

- *Page 1648, Lines 2 and 3: Replace 'soluble' with 'solubility' (x2)*

Answer: Changed

- *Page 1648, Lines 5-7: Remove these sentences. They are repetitive as they are used in the figure legend as well as the proceeding section.*

Answer: Sentences are removed.

- *Page 1649, Line 10: Replace 'soluble' with 'solubility'. This appears to happen throughout the text – please check that all usages are correct.*

Answer: Changed

- *Page 1652, Lines 5-6: Change 'the facility which leads off waves being reflected to the walls, results to a different wave field than on the open ocean' to 'the facility, which leads to waves reflecting off the walls and results in a different wave field to that found in the open ocean'*

Answer: Changed

- *Page 1652, Line 16-21: Most of this information is repeated in the subsequent air and water phase sections. To reduce duplication of information, please remove this para.*

Answer: Here, we introduce the instrumentation used for the concentration measurements. Full names and providers are given for both instruments while in the following sections only the abbreviations are used. The authors wish to maintain the structure of the manuscript and keep these introductory sentences.

- *Page 1653, Line 4: No need to discuss the measurement approach for halocarbons. Please remove.*

Answer: Please see answer to the general comment at the beginning of this review. Since this work forms the methodic base to our follow-up analysis paper we believe it is important to describe the experiment in its entirety.

- *Page 1652, Line 27: Insert 'air and water' between 'both' and 'phases'.*

Answer: Changed

- *Page 1653, Line 13-16: This sentence is too long, confusing and poorly worded. Suggest you divide into two sentences and rephrase.*

Answer: Indeed the sentence is confusing. Text is corrected accordingly.

- *Page 1654, Line 2: Replace 'avoiding' with 'to avoid'*

Answer: Changed

- *Page 1654, Line 17: Replace 'to a field' with 'in a field'*

Answer: Text has been removed. See comment of referee 1.

- *Page 1660, Lines 22-24: The sentence beginning 'For the last condition..' would be more appropriate in the Table legend. It is also unclear and needs to be reworded. I suggest 'For the highest wind speed condition of the clean case, only three repetitions were performed.'*

Answer: Text has been changed as suggested.

- *Page 1660, Lines 24-25: This sentence is not very clear and initially seems to be a poor justification for subjectively removing data. The subsequent sentence attempts to justify removal by suggesting that the driver is a insufficiently skimmed water surface. If so, this would appear to be a source of potential error in the measurements. It suggests that the authors should exclude all of the data from repetition 2 rather than just the mean square slope.*

Answer: We do not present our k_t results as an average of 4 repetitions. We show each k_t value to its correlated $u_{*,w}$ (see Figs. 7 and 8). In table 2, we aim to provide the reader with an overview of the conditions used. We use average values as the detailed table would be very big, containing repeated information. In cases where a repetition for some reason behaved as an 'outliner', the data were not used for averaging. The data though, are still used in our plots and they are very interesting as they might provide a better understanding of the effect of each parameter on the gas exchange. For example see comment in Section 4.1 second paragraph.
For this reason, the authors would like to keep this sentence.

- *Page 1660, Line 29 – Page 1661, Line1: See my general comment about interpolating u* between the high surfactant and clean cases.*

Answer: Sentences have been removed as now the new $u_{*,w}$ data are used, where all surfactant cases have been measured.

- *Page 1661, Lines 5-15: See earlier comments about 14 tracers. This paragraph seems unnecessary.*

Answer: The first two sentences of the paragraph have been removed. Paragraph has been rewritten. (See comment of referee 1.)

- *Page 1661, Lines 18-22: The sentences beginning 'In both figures: : :' and 'Vertical light bars: : :' should be in the figure legend, not the main text.*

Answer: Sentences have been placed in the figure legend.

- *Page 1662, Lines 5-6: This sentence beginning 'Small variations: : :' does not make sense.*

Answer: Sentence has been rewritten.

- *Page 1662, Lines 8-9: Given that repetition 2 gave lower mean square slope values, I think the data should be plotted against more than one physical parameter. Further, as the authors themselves note, 'one physical parameter [may not be] enough' to describe the processes controlling gas transfer. For these reasons, the gas transfer data should be plotted against Uref as well as against u*. In many ways this would be more useful information than the inset log-log plots in Figs 7 and 8.*

Answer: After analyzing the transfer velocity data, it became clear that the $u_{*,w}$ is best parameter to use in the plots. It is also the most common one (together with u_{10}). Plotting the data against u_{ref} would not provide any new information. The $u_{*,w}$ is calculated after the u_{ref} (u_{ref} - $u_{*,w}$ equation) . The reference wind speed, u_{ref} is correlated to Aeolotron facility and cannot be compared with previous studies as is the case for the $u_{*,w}$.
The comment here suggests that both ($u_{*,w}$ and mss) parameters seem to be significant. How significant each of them is and in which way both of these parameters could be combined to describe the gas exchange process is not the purpose of the current publication and it will be presented in the follow up paper.

- *Page 1663, Line 4: change 'decrease' to 'suppressed'*

Answer: Changed

- *Page 1663, Lines 6-8: The sentence beginning 'The clean water surface: : :' belongs in the legend of Fig. 9*

Answer: Sentence has been placed in the legend of Figure 9.

- *Page 1663, Line 11: I disagree with the word 'ineffective'. Suggest you replace with 'less effective'.*

Answer: Word has been replaced.

- *Page 1664, Line 17: Replace 'weakly soluble, high soluble' with 'weakly soluble gases, high solubility'*

Answer: Corrected

- *Page 1664, Line 27: Remove comma*

Answer: Corrected

- *Page 1665, Line 2: Replace 'deriving' with 'derived'*

Answer: corrected

- *Page 1665, Line 3: Please rephrase this sentence to make clear which studied 'these studies' refers to.*

Answer: Sentence is rephrased.

- *Page 1665, Line 13: Remove comma.*

Answer: Corrected

Figures and Tables:

- *Table 1: See my general comment. I cannot see why a study that only presents data from two gases should include any information about the other 12 gases. Please remove this table.*

Answer: Please refer to the answer given to the general comment at the beginning of this review. (We need this table for this method paper to be useful for the follow up analysis paper.)

- *Table 2: This table requires some further explanation as it confused me initially. Pleas rename the 'conditions' with something more meaningful. I suggest using either the relevant Uref or U*w. In the legend, insert the word 'Case' before '1', '2' and '3' to make it clear that the cases are the clean vs Triton experiments. Also, you need to state somewhere that the multiples in brackets refer to the number of replicates performed for each Case.*

Answer:
Table 2 without the u_{ref} values would indeed look simpler to understand. Though, the u_{ref} values are absolutely necessary as a reference point. As it is also mentioned in the text, the $u_{*,w}$ and mss values are affected by the surfactant. The effect in each case can be approximately interpreted taking the u_{ref} as a reference point and comparing $u_{*,w}$ and mss with the clean case.

Rest corrections have been included in the table. A clarification for the word 'cond.' is added in the legend.

- *Figure 2: Configuration makes it difficult to understand. Suggest configuration is changed so that it is a 3x2 set of panels. The panel a (wind speed) can be repeated in the top left and top right panels. Then the left hand column would be air and water concentrations for waterside experiments and the right-hand column would be the same for air-side controlled experiments.*

Answer: The authors submitted Figure 2 in a configuration where the 5 panels fit the one under the other. This configuration will appear in the final version of this publication, so it would be easy to follow. The split of panels was necessary for the discussion version only.

- *Figures 9-11: These data need to be plotted with the transfer velocities not log transformed if we are to really see how well the data compare with previous results and the magnitude of the surfactant effects on waterside controlled gas transfer.*

Answer: The authors believe that a log-log representation of the results is necessary in order to better present the wind speed regimes at which the data agree with the previous studies and the regimes where they do not agree. This is only possible using the log-log scaling. In a linear scaling, only the differences reflecting the high wind speeds are to be noticed where, as also mentioned in the text, the biggest discrepancies are apparent at low wind speeds (especially in Figure 10). Since, both referees wish to see Figure 10 and 11 in a linear scale, these have been introduced additionally.