Response to Referee #1
"Ocean bubbles under high wind conditions. Part 2: Bubble size distributions and implications for models of bubble dynamics" by Helen Czerski et al., Ocean Sci. Discuss. https://os.copernicus.org/preprints/os-2021-104, 2021
March 2022

We are grateful to the reviewer for their comments and have prepared a revised manuscript which addresses their concerns. Our response to each point and a description of the changes made are given below (reviewer comment in blue, author response in black).

The paper presents results from the field campaign highwings reporting on the bubble measurements in the first 10meters below the sea surface using optical and acoustic techniques. The paper is important for the community as very few of such measurements exist. I have a set of recommendations or suggestions for the authors to improve the manuscript.

In particular, I strongly recommend to use the published data from highwings on gas transfer, waves and whitecap coverage and see the correlation with the bubble plume. The data are available online and have been used by various authors . The bubble plume measurements are by construction fragmentary since the full entrainment process is not captured but it is certainly a very valuable/important information. Combining this information together with the gas transfer velocity, the whitecap coverage and comparing with recent models and parameterization would shed light on the transport process of the bubbles and how the present data can be used to better constrain these models.

We understand the desire to see an analysis of bubble data in relation to air-sea gas transfer data. We are well-aware that the data exist – two of the authors of this paper are also co-authors on the HiWINGS gas transfer publications. We do not present that analysis here for these reasons:

- Firstly, we note that we do make comparisons with wave parameters, both in the companion paper (referenced multiple times in this paper) and in Appendix C of this paper, where we show that the bubble data does not show clear patterns when segregated by the wind-wave Reynolds number or the significant wave height.
- The bubbles produced by breaking waves are important for several different fields: aerosol production, optics and underwater acoustics as well as air-sea gas transfer. Our aim here is to present basic data on bubble size distributions which is useful to all the fields that need it, rather than bias the presentation by tuning it to a particular question. There is a severe lack of basic information about the composition of deep bubble plumes, and addressing this is the priority.
- A follow-on study could indeed compare the air-sea gas transfer and the bubble parameters. However, this is a significant separate piece of work, and this paper (which, as noted above, covers extensive details about bubble plumes which have not previously been observed) is already close to 12,000 words long. We do not consider that the community would be well-served by us removing details of the basic data analysis in order to make room for gas transfer comparisons. In short, a gas transfer

paper would be desirable but would be presented in a different paper. We also note that it would be based on the fundamental observations presented here, which necessarily come first. We are seeking the resources to do that additional study, although since all the data is public others could also take on that task.

- Our bubble data was measured at depths of 2m and below, and it is generally accepted that the majority of CO_2 uptake occurs very close to the surface. As we state in the paper, we think that a full understanding requires measurement of the flow processes and the dissolved gas spatial distribution in addition to the bubbles, and we think that a simplistic comparison that ignores those complications is unhelpful.
- We also note that there are extensive assumptions made in modelling studies about the structure, composition and movement of bubble plumes, but very little available data to base those assumptions on. We therefore consider that a first step towards helping the modelling community is in providing constraints from field data, which can be used to modify and validate their models.

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In the introduction, you state "If normalised by void fraction, these distributions collapse to a very narrow range, implying that the bubble population is relatively stable and the void fraction is determined by bubbles spreading out in space rather than changing their size over time." Back of the envelope calculations of bubble mediated gas exchange can tell you that the transfer of CO_2, O_2 is relatively slow compared to your observation time of the bubbles, so that bubbles will not change size significantly in the first few meters even if they are exchanging gas (very true for CO_2 which does not account for much of the volume). Bubbles will only change size if they are brought deeper in the flow by some turbulence process (Langmuir turbulence, etc). This is discussed in the earlier papers from Keeling 1993, Woolf and Thorpe and is implicit in Liang et al 2011, 2012 or Woolf' modeling. I would recommend looking at recent modeling work on that topic:

Leighton TG, Coles DG, Srokosz M, White PR, Woolf DK. 2018. Asymmetric transfer of CO_2 across a broken sea surface. *Sci. Rep.* 8:8301

Liang J-H, McWilliams JC, Sullivan PP, Baschek B. 2011. Modeling bubbles and dissolved gases in the ocean. *J. Geophys. Res.* 116:C03015

Liang J-H, McWilliams JC, Sullivan PP, Baschek B. 2012. Large eddy simulation of the bubbly ocean: new insights on subsurface bubble distribution and bubble-mediated gas transfer. *J. Geophys. Res.* 117:C04002.

We are aware of <u>no</u> direct in-situ measurements of bubbles changing radius through dissolution in the top few metres of the ocean, and only one attempt (Czerski, 2011) to directly characterise the bubble coating in the open ocean. The parameters at play are understood - bubble radius, surfactant coatings, the possible contribution of particulates to stabilisation, pressure and the local gas saturation – but there is no actual data from the ocean to tell us whether the bubble models currently used balance these influences in the right way. Our data set is valuable precisely because it includes the most detailed measurements of bubble size distributions in high winds to date, and this can be used to test models. Our approach here is to avoid assumptions, and to base our analysis on field data (ours and that of other people) only. So we think it important to make statements like the one the reviewer is commenting on because this is what actual measurements imply, unclouded by the assumptions often made for the purposes of modeling. If our data support the models, the point is not that we are making comments that lack novelty, but that when faced with actual field data, the models match the measured reality.

We have looked at the three papers suggested by the reviewer.

The Leighton 2018 paper discusses models which are based on minimal data, with significant limitations in the data collection technique that are not addressed or discussed (either in the main text or the supplementary material). The data that we present from our study is significantly more robust, with methods and the limitations from environmental conditions carefully described. We do not see any reason to re-interpret our results based on this 2018 study, when it is far more limited in scope and quality than our own.

The Liang 2011 and 2012 papers develop a bubble model which presents a similar overall picture to our data, and we are grateful to the reviewer for bringing these to our attention. However, we note that they are not directly tested against field data except in a general way, and there are places where the model outputs do not agree with our data (which is perhaps not unexpected considering the breadth of the model and the limited field data available when they were written). We have added these papers to our literature review, and we have also added a brief comment on the comparison between their data and ours at line 558.

Towards the end of the intro, you state: "We suggest that as bubbles move to depths greater than 2 m, sudden collapse may be more significant as a bubble destruction mechanism than slow dissolution, especially in regions of high void fraction." Yes, this is discussed by modeling studies from Liang et al and Woolf et al. The Langmuir type entrainment process is necessary to bring small bubbles down where they can collapse due to hydrostatic pressure. However, the life of the larger bubbles in the top two meter is important for co2 transfer, they exchange gas during their lifetime without changing the bubble size, since the content of CO2 is small compared to the overall volume of the bubble. This is discussed or implied in the models by Keeling 1993 and then Deike and Melville 2018

As we emphasize above, our aim here is to present experimental data by itself without relying on the interpretations that have come from modelling. We think that it is important to be clear about the implications of our data, and we note that one of the other referees thought that collapse was a novel concept that required strong additional justification, suggesting that this possibility is far from accepted. We are not making claims to novelty here; we are stating the conclusions from our own field data. We also note that the word "collapse" does not, as far as we are aware, appear in any of the papers the reviewer mentioned, except when used to describe the collapse of data sets on to a single line, or for the deformation of large air pockets during the breaking process. It may be implied and obvious to those creating the models, but not apparent to others. We are talking about a sudden process happening at depth, and we were not aware of this suggestion being made explicitly elsewhere in the literature. We want the language we use here to be clear and unambiguous.

I would recommend citing Bowyer PA. 2001. Video measurements of near-surface bubble spectra. *J. Geophys. Res. Oceans* 106(C7):14179– 90; which present of the few direct optical measurement of bubbles below the ocean surface.

We have added this citation on line 80.

I do not understand the statement: "Our results suggest that local gas supersaturation around the bubble plume may have a strong influence on bubble lifetime, but significantly, the deep plumes themselves cannot be responsible for this supersaturation"

In conversation with other researchers in this field, we have noticed a strong tendency to assume that the deep plumes are significant because the gas contained in those bubbles will be injected into the ocean at depth and will significantly increase the gas saturation in the water at that depth. Our point here is that it seems unlikely that there is enough gas in the bubbles in the deep plumes to make a significant difference, but we think that the bubbles may continue to exist because they are in a water packet with higher than average gas saturation. We are suggesting that the main gas contribution associated with a deep plume is already in the dissolved phase, rather than the gaseous phase. This is explained further in the main text at line 425. We have changed the phrasing in the abstract to clarify our meaning and it now reads "… *significantly, the gas in the bubbles contained in the deep plumes themselves cannot be responsible for*…". (line 30).

The authors present data on bubble void fraction, distribution, etc. During the same campaign, the gas transfer velocity (or piston velocity) for CO2 and DMS has been measured and is reported in Brumer et al 2017 (GRL) (which is not cited), as well as whitecap coverage data. Could the authors present cross comparison of these quantities? Similarly, Deike et al 2017, and then Deike and Melville 2018 presented scaling for air entrainment by breaking wave and some of that has been used to predict gas exchange. While your data do not get the full air entrainment because you are missing the large bubbles close to the surface it would be interesting to see whether the proposed scaling in the literature in terms of dependency with wind and wave can work or not. Similarly, Brumer et al 2017 proposed a wave Reynolds number scaling for gas transfer and it would be interesting to see if your bubble data follow such scaling. Finally seeing the correlation between whitecap coverage data from Brumer et al 2017 and your bubble data would provide information on how much of the bubbles are being transported down to the depth where you are making the measurements. This could be very useful for future modeling on the role of small bubbles getting fully dissolved in the water column, which will contribute to exchange of less soluble gases such as O2, N2.

Brumer S, Zappa C, Blomquist B, Fairall C, Cifuentes-Lorenzen A, et al. 2017a. Wave-related Reynolds num- ber parameterizations of CO2 and DMS transfer velocities. *Geophys. Res. Lett.* 44(19):9865–75

Brumer SE, Zappa CJ, Brooks IM, Tamura H, Brown SM, et al. 2017b. Whitecap coverage dependence on wind and wave statistics as observed during SO GasEx and HiWinGS. *J. Phys. Oceanogr.* 47(9):2211–35

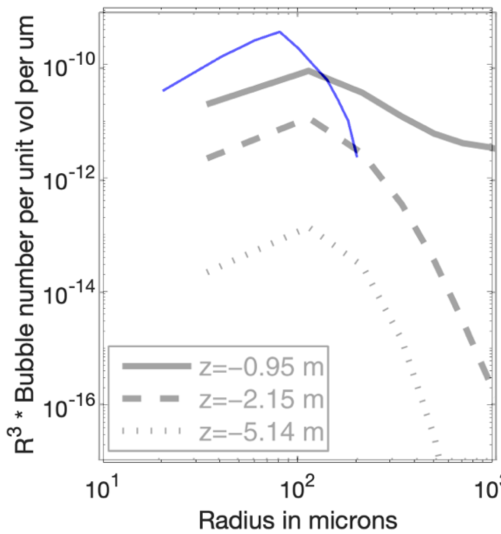
Deike L, Lenain L, Melville WK. 2017. Air entrainment by breaking waves. *Geophys. Res. Lett.* 44(8):3779– 87

Deike L, Melville WK. 2018. Gas transfer by breaking waves. *Geophys. Res. Lett.* 45(19):10–482

We are well aware of the gas transfer data from this cruise, as stated above in our response to the general points. We have provided our reasons for not making extensive comparisons to the gas transfer data there: that is a separate piece of work. This is a more fundamental paper dealing with the bubble measurements, and any gas transfer comparison would build on the work presented here.

About figure 2: the shapes at 2m seem compatible with the modeling work from Liang et al; which starts from a Deane and Stokes like distribution - similar to other recent bubble gas transfer formulation. While a quantitative comparison is probably out of scope, this should be mentioned. It seems that the present measurements are compatible with the current assumptions from bubble models.

We assume that the reviewer is referring to figure 6 in Liang 2012, since this is the only plot we can find that refers to a depth of 2m, although the wind speed for those models is only 10 m/s. According to the data in our companion paper, the dominant void fraction at 10 m/s wind speed and a depth of 2m is 10^{-7}, so the relevant comparison is the bubble size distribution shown in blue and turquoise lines in figure 4(a) of this paper. We have taken the $10^{-7.5} – 10^{-7}$ void fraction size distribution for this wind speed range, multiplied each point by the radius cubed, and to the left we show the overlay of the two plots. The comparison is between the 2.15 m dashed line and the blue one.



The discrepancy here is about a factor of 100, although the shape is similar. This is clearly a relatively crude comparison, but it seems a stretch to say that the model is compatible with the data. We have not added a comment to the paper, since this comparison is limited but does not suggest that the model outcomes are appropriate in this case.

Similarly and about all figures on bubble size distributions. Can you plot your integrated volume from these distribution as a function of wind speed? void fraction? wave age? wave height? Whitecap coverage? Gas transfer velocity? This could be useful to compare with existing models from air entrainment based on breaking dynamics (assuming the air lost between entrainment and 2m down scale in a similar way which is not obvious at all). This would provide very useful information/constraints for modeling.

The companion paper (now accepted for this journal, https://doi.org/10.5194/os-2021-103) focusses on void fraction distributions and how they change with wind speed, wave age etc. This data is all presented there (with the exception of gas transfer comparisons, for the reasons stated above).