Response to second round of reviewer comments of Quantifying the vertical transport of $CHBr_3$ and CH_2Br_2 over the Western Pacific by Butler et al

We thank again the reviewer for these further comments. We have addressed each reviewer comment (denoted by italics) and changed the manuscript where appropriate. We apologize for the delay in our responses.

P1, L10: "In the absence of reliable ocean emission estimates, …". If this statement is correct, then none of the tagged simulations would've have any sense. So please be careful when specifically justifying your work, specially within the abstract and conclusions. You may rather replace "reliable" by "high resolution estimate" or "local estimate".

We revised this statement to reflect this comment "In the absence of local ocean emission estimates…"

P1, L9: "… and by older air masses that originate upwind". What do you mean by upwind? That bromine sources are somehow generated above the surface? (See related comment below).

They originate from the region outside of our study region. We have clarified this in the manuscript by stating "…but it is still dominated by emissions from the open ocean and by older air masses that originate outside of our study region."

P2, L29: Fernandez et al., 2014 has also provided estimates of PGI and SGI contributions lying within this range.

We have now included this reference.

P3, L5: Specific clarification of using only SG measurements from CONTRAST and CAST should be given, as those campaigns also measured PGs in the UT.

We have now changed this statement to "We use source gas data from two coordinated aircraft campaigns…"

Section 2.1. Provide specific information of the exact dates when the campaign was performed.

We have added dates to the opening sentence in Section 2.1: "We use CHBr3 and CH2Br2 mole fractions from the CAST and CONTRAST aircraft campaigns, running from 18/01/2014-28/02/2014"

Section 2.2. This section only points to Table 2 (which only shows the locations of NOAA stations) and then points to Appendix A-1. I suggest moving the whole NOAA validation section to the Appendix, including Tables 2 and 3, and summarize within a paragraph the main results in the text.

We have now moved Table 3 to the appendix. In Section 4.1 we have left a short paragraph summarising results from the NOAA evaluation.

Section 3: When the model description is given, no reference regarding the period of time modelled is provided. The latitude/longitude limits used to define the western pacific region are not defined.

We thank the reviewer for spotting this oversight. We have modified the text accordingly:
"We initiate the model on 1 January 2014 and run until 1 March 2014. The Western Pacific region is defined as 120°—170°E and 30°N—20°S. This encompasses the full region covered by the CAST and CONTRAST measurements." In the model description.

P5,L7: "Figure 2 shows the magnitude and spatial distribution of our prior emissions of CHBr3 and CH2Br2 (Liang et al., 2010)." What do you mean by "prior"? You have not modified them into a top-down like approach to adjust the Liang emission to your model. So the emissions are kept constant throughout the whole study. Also, indicate if the Liang inventory includes any "coast-to-ocean" scaling factor that could affect the results and conclusions from your work?

The term 'prior' is a slight misuse of language. This has been changed to "Figure 2 shows the magnitude and spatial distribution of the $CHBr_3$ and CH_2Br_2 emissions (Liang et al., 2010)."

P5,L9: Reported emissions from Liang et al, 2010 are not 396 Gg Br yr−1, but 425 Gg Br yr−1. Please check.

We thank the reviewer for spotting this typo.

P5,L14: "We chose to use Liang et al. (2010) because it has a consistent bias for CHBr3 and CH_2Br_2." I can imagine that you can find a better reason for choosing the Liang inventory than this one. Also, is it the ocean tagged version identical to the Liang emissions? (within the WP region)

The reviewer is right that we could have used a number of arbitrary criteria to select an emission inventory, but we chose Liang because of we found a consistent mean bias of these two gases. This in turn allows us to confidently analyse the ratio of these two gases in a complementary paper (Feng et al, https://doi.org/10.5194/acp-2017-949). We confirm that the ocean tagged region represent the Liang emissions. For VSL sources, we are limited from the number that we could choose.

P6,L9: Rx has not been defined

We thank the reviewer to spotting this error. We have changed the text to read "Fractional contributions of tracers are calculated based on relative ratio of each tracer within a grid box (Rx)…"

P7, L11: If the formula for bias computation is included in the main text, then it should be explained. I suggest just moving to the figure caption.

Agreed. The bias calculation is now in the caption of Figure 4.

P7, L3: "Model errors in reproducing the observed seasonal cycle reflect errors in production and loss rates." How do you compute "production rates" from SGs? Do you mean "errors in VSL sources and loss rates"? All VSL chemical mechanism I am aware of include only decomposition of VSL halocarbons by reaction with OH and hv, so you can compute the loss rates. But there are not any VSL production rates due to gas-phase reactions, only emission of source gases from the ocean.

Agreed. We have this changed this to "emission and loss rates."

P7, L9-10: "Larger differences in the correlations for CH2Br2 is likely due to differences in the sampled air masses that have originated far upwind." Once again, what do you mean by upwind? You should make this explanation clear in the text. Also, please relate the SGs surface analysis to the TransCom-VSL paper (Hossaini et al., 2016) and their findings respect to the global model performance in reproducing VSL SGs in the surface and UT when different emission scenarios are used.

We agree this is a difficult statement to understand. We have now changed the text to:

A recent model inter-comparison showed that different combinations of models and prior emission inventories resulted in large variations of surface model concentrations at station sites [Hossaini et al, 2016]. This study that the Ziska inventory was most consistent with observations but not for all models. Model agreement with UT observations was generally better but still showed large inter-model variations particularly over the Western Pacific where there is a strong convective transport.

Later in P8, L10-12, the authors cite the TransCom-VSL paper, but they seem to be pointing out to how different models behave differently when different emissions are used, while the TransCom-VSL paper highlights that most global models used were capable of reproducing VSL SG in the TTL independently of the emission inventory used.

Hoissani et al report that
"Overall, model–measurement agreement of CHBr3 in the TTL is poorer during the ATTREX campaigns, with most models exhibiting a low bias between 14 and 16 km altitude. MOZART and UKCA simulations (which prefer the Liang CHBr3 inventory) exhibit larger mixing ratios in the TTL, though are generally consistent with other models around the tropopause. Most (≥ 70 %) of the models reproduce CHBr3 at the tropopause to within ±1σ of the observed mean and all the models are within the measured range (not shown) during both ATTREX campaigns. Model–measurement CHBr3 correlation is > 0.8 for each ATTREX campaign, showing that again much of the observed variability throughout the CHBr3 profiles is captured."

We agree that transport processes are key in understanding VSL SG in the TTL, but there *is* a role for emission inventories.

P7, L5: I understand the intention of the authors, but I do not see the inverted S shape in the VSL vertical profile in Figure 4. Also in P8, L3 and elsewhere, the inverted S shape is mentioned but is never explained nor justified. Which are the processes producing this observed feature?. (See my comment on Fig. 4 below).

We have addressed this comment below when the reviewer raises the issues of better labelling.

P7, L27: "Prevailing easterly transport of gases over the region is dominated by the vast area of open ocean sources that appear to weaken the magnitude of spatially limited coastal emissions (Andrews et al., 2016; Pan et al., 2016)." How can the open ocean "weaken" the coastal emissions?" Please rephrase and explain.

We have rephrased this to "Prevailing easterly transport of gases over the region is dominated by the vast area of open ocean sources that appear to dominate the magnitude of spatially limited coastal emissions (Andrews et al., 2016; Pan et al., 2016)."

P7, L30: It would be very useful to compare the percentage contribution of coastal emissions to CH_2Br_2 in the TTL respect to the percentage contribution of $CHBr_3$, and explain it in relation

of their predominant sources and lifetimes. Also perform the same comparison for percentage contribution of the open ocean tracers to each species in the TTL.

With respect for the sake of readability, we do not understand how this requested change would add to the paper as it is now, especially given its current length. We already show how much of each gas enters the TTL from individual ocean sources.

P7, L33: "with the remainder originating from emissions prior to the campaigns.". How are you capable to distinguish that the reminder is from emissions prior to the campaign and not from sources located outside of the tagged tracers?

We define "Background conditions are representative of atmospheric concentrations before the campaign started as they do not include emissions during the campaign period."

P8, L2: "…with contributions from geographical regions immediately outside the study región…". How do you know they are "immediately" outside? Have you performed an additional tagged simulation with emissions from the surrounding areas of the WP? How do you relate this statement with your previous comment regarding the emissions originated prior to the campaign (and not outside of the WP)?

This is not a conclusion we can reach with the experiments we have completed. Consequently, we have changed the statement to "Coastal ocean emissions represent a smaller contribution to $CHBr_3$ at lower altitudes, but increase their influence above 6 km in the CONTRAST data reaching a maximum of 60% of the total CHBr3 tracer in the TTL."

P8, L6-9: I found confusing that the "largest" contribution reach a maximum of 28% and that the "reminder" (e.g., 72%) is not referred as the dominant contribution.

We have now emphasized that these are the largest contributions of emissions during the campaign period. "The ocean, in particular the open ocean, represents the largest contributions to total CH_2Br_2 of emissions during the campaign period. They typically represent 20% of the total CH_2Br_2 and reaching a maximum of 28% in the TTL for the CONTRAST measurements. Maximum contributions of coastal emission sources peak at 15% of total CH2Br2 tracer in the TTL, much less than for CHBr3. The remaining contributions (72%) are representative of the emissions prior to the campaign period."

P8, L31: "despite intensive measurements around coastal land masses of the region". What do you mean by coastal land masses?

We mean islands. We have revised this statement "despite intensive measurements around coastal areas of the region".

Figure 8: (P8, L15): Why coastal seems not to contribute to the total ocean profile, which seems to be very similar to the open ocean. I would expect the total to be the sum of both coastal and open ocean profiles.

This is representative of the relative strength of each of the emission source regions. The weak emission source region of the coastal ocean will not contribute to the age profile. Due to the smaller area coverage of the open ocean, it will be weakened compared to the total ocean age profile.

P8, L23-26: I was surprised that the coastal tracer percentage contribution to the TTL was found to be much smaller than the open ocean contribution. Could you provide some insights on this interesting result? Is it because of the assumed source distribution? Is due to the different transport regimes and speed of convection?

This is in relation to the weak strength of coastal ocean emissions. Coastal ocean sources have a relatively smaller source distribution compared to open and total ocean, likely to lead it to have weaker strength in being transported to the TTL.

P8, L32: Deficiencies could also be due to an incorrect representation of the spatial distribution of VSL sources in the inventory used (you mentioned at other places of the text).

The age of air calculation is based on bathymetry data and not VSL sources. This is high resolution data that has been averaged over the model resolution, this model resolution will therefore be the dominant cause of mismatched results.

Figure 10 (P8, L34, P9, L3): I do not understand what the intention of including Fig. 10 is nor the analysis performed here. Please describe it in more detail or remove it. Other reviewer also highlighted this issue during the first round of review.

We have clarified this text in response to this comment by emphasizing our key points that a) although CH3Br values appear to be insensitive to age they are in fact the superposition of slow ascension of higher coastal emissions and the faster ascension of lower open ocean emissions. We believe that is a useful observation to include in the paper.

"Figure 10 shows mixing ratios of $CHBr_3$ decreasing with altitude, but remaining fairly constant with increasing age within each altitude range. We find that $CHBr_3$ values are determined mainly by younger air masses from the open ocean and older air masses by coastal emissions (Figure \ref{fig:freq_db_age}). Coastal emissions are associated with the highest surface emissions but they also subjected to slow ascent rates and consequently greater photochemical losses. In contrast, open ocean emissions are lower than coastal emissions but are convected more rapidly and subject to less chemical loss. Consequently, $CHBr_3$ appears to be insensitive to age but is in fact a superposition of young and old airmasses from different origins. From our analysis, we found that $CHBr_3$ values are determined mainly by younger air masses from the open ocean (Figure 8). Within the TTL, higher median mole fractions are associated with the highest model convective mass flux in each age bin. The peak frequency for the mean age of air in the TTL is 48–72 days, corresponding to $3\tau_{CHBr_3}$ and median values of 0.5 pptv $CHBr_3$ from oceanic emission sources, and 0.6 pptv in high convective systems. However, less than 0.5% (2%) of air being transported to the TTL within 24–48 (48–72) days of emission are associated with high convection events. Weaker, mean convection plays an important role in more consistently transporting large mole fractions to the free troposphere that is then transported more slowly to the TTL. "

P9, L18-34: I believe that in the discussion a comparison with the results obtained from Navarro et al., 2015 during ATTREX in the same region of study should be given. Also, relate your results to other papers reporting CONTRAST and/or CAST data.

This is already included in the revised manuscript. "This is consistent with \cite{Navarro2015} who estimate VSLS contribution over the Pacific from observations in 2013 and 2014. It estimates 3.27$\pm$0.47~pptv of bromine from CHBr$_3$, CH$_2$Br$_2$ and other minor VSLS sources at the tropopause level (17~km)."

Section 5: There is no discussion at all, only a summary of the results previously presented. So it should only be called "Concluding Remarks".

Point well taken. Section 5 is now titled “Concluding Remarks”.

P10, L4-5: “…due to advection of air masses convected from areas outside the study region…”. Is this contribution dependent on the strength of convection or on the large scale ascent?

“Coastal ocean sources typically contribute 20\% to total atmospheric CHBr$_3$ but reach a maximum of 60\% in the TTL due to advection of air masses convected from areas outside the study region due to large-scale ascent.”

P10, L9: “…are dominated by sources from before the campaign.” Here in the conclusion the “reminder” contribution seems to be the dominant source. Once again, the authors need to explain how the contribution from “before” the campaign and from “outside” the region are recognized and distinguished.

Agreed. “are dominated by sources emitted outside the study period.” Our tagged approach that quantifies this source contribution is described in section 3.1.

Figure 4: You should use a,b,c,d,e,f labels for each independent panel. In panels a.b (top row) I suggest using filled colored boxes for model output and empty colored boxes for campaign data. Also explain that the model and observations data corresponding to the same altitude interval are slightly shifted in the vertical axes for each bin.

We have now labelled each independent panel and used coloured boxes for the model panels. We have also added a mean vertical profile show the backwards ‘S’ profile shape we describe in the main paper.

Figure 8: Indicate in the figure caption what the vertical dashed lines indicate.

I have added the meaning to the figure caption: “One e-folding lifetime of CHBr3 of 24 days (vertical dashed line) and CH2Br2 of 123 days (vertical dotted line) are indicated.”

Figure A-2: The sigma errors are denoted by vertical (not horizontal) lines.

Thank you for spotting the error. This has been corrected.

Appendix A: The information given in the last paragraph could be moved as a summary of the NOAA validation into the main text. Table 3 should be in the appendix.

Table 3 has been moved in to the appendix.

P29, L4-5: “This variability will represent the large variability of convective events over the region, as well as the aforementioned errors in model emissions.” This sentence is confusing and out of context in the appendix. Note that the appendix should be read as an independent portion of the work.

This sentence is in the appendix at the request of a previous review. But we have since decided to delete it.