

2nd Review of “Quantifying the vertical transport of CHBr₃ and CH₂Br₂ over the Western Pacific” by Robyn Butler et al.

Many improvements have been achieved since the 1st round of review. The authors have properly addressed the specific issues I raised before (Specific comments I, II and III), being the most important the correction of including an independent land tracer for VSL emissions. However, I still find that the description and discussion of the results can be greatly improved, and that the main results found can be written in a more rigorous fashion. Indeed, my general concern during 1st round of review was: “... and most importantly, I found many of the analysis and discussions given in Sections 4 (Results) and 5 (Discussions and Concluding remarks) very vague and/or requiring stronger evidence that supports them. I suggest being more specific and formal on the explanation of the specific statements highlighted in this review”; which was not answered nor commented in the response letter.

I found that many of my original issues raised in the review were properly answered in the response letter. But when I read the 2nd draft, those answers were not so clearly described or justified in the manuscript text. For example, the answer letter points out that (P2) “... e.g., coastal emissions play a larger role at higher altitudes.” or (P7) “...The total tracer is based on a 12-month spin up from the emission scenario with the ‘total ocean’ emissions included. The VSLs VMR that is not represented by the tagged tracers will be attributed to ‘background’ VMR from the spin up file, which represents the VMR from before the campaign. We have clarified this in the method and discussion sections”. I could not find those (or equivalent) sentences in the manuscript. Please, be sure to include all comments from the response letter into the manuscript.

As I previously mentioned, “I found the study very interesting and of relevance for ACP”, but I believe redaction/quality improvements can be achieved before final publication. Those should be oriented to avoid using misleading interpretations if individual sentences are extracted from the paragraph (P1, “In the absence of reliable ocean emission estimates,...”; or P5, “We chose to use Liang et al. (2010) because it has a consistent bias for CHBr₃ and CH₂Br₂.”) and to justify the contributions of other factors/scenarios not explicitly considered in the tagged experiments performed in this work (P7, “Larger differences in the correlations for CH₂Br₂ is likely due to differences in the sampled air masses that have originated far upwind.”). Additionally, the manuscript would benefit of a comprehensive discussion on how the Butler et al results relate to previously published papers already in the literature (mainly those performed in the same region of study). Below I point out to the main phrases/sentences in the text where I found some ambiguity or confusion that should be improved.

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".

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).

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

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.

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

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.

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.

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?

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

P5,L14: *"We chose to use Liang et al. (2010) because it has a consistent bias for CHBr3 and CH2Br2."* 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)

P6,L9: Rx has not been defined

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.

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.

P7, L9-10: *“Larger differences in the correlations for CH₂Br₂ is likely due to differences in the 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.

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.

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).

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.

P7, L30: It would be very useful to compare the percentage contribution of coastal emissions to CH₂Br₂ in the TTL respect to the percentage contribution of CHBr₃, 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.

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

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)?

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

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

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.

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?

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).

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.

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.

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

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.

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.

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

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

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.

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.