Interactive comment on "Transport of short-lived halocarbons to the stratosphere over the Pacific Ocean" by Michal T. Filus et al.

Anonymous Referee #1

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This work builds on the 2014 joint CAST/CONTRAST/ATTREX missions where VSLS ($CHBr_3$, CH_2Br_2, CH_3I) measurements were made in the tropical West Pacific. Here, the NAME model is used to compute back trajectories from the VSLS measurement location/times and to determine the fraction of released particles that crossed the boundary layer in the preceding 12 days. With this information, the authors estimate the influence of the boundary layer on VSLS mixing ratios (during the campaign period) throughout the vertical extent of the TTL on differences in measured VSLS concentrations between ATTREX 2013 (W Pacific) and 2014 (E Pacific).

My main concerns (outlined below) are on the use of assumed chemical decay times and on some aspects of the manuscript presentation. Both issues could be addressed readily, and I recommend the paper for publication.

We thank both reviewers for their constructive comments. In our opinion, these have resulted in an improved manuscript.

(1) The authors use constant chemical decay lifetimes of 15 days and 94 days for $CHBr_3$ and CH_2Br_2, respectively, based on the boundary layer estimates given by Carpenter et al. (2014). Can the use of a fixed lifetime be justified given that local lifetimes of the above compounds are known to vary substantially between the surface and in the TTL (e.g. Hossaini et al., 2010, Liang et al., 2010)? These references show a much longer TTL CH_2Br_2 lifetime than 94 days, for example. Accounting for photochemical removal along trajectories is important and the authors should comment on how sensitive their findings (e.g. boundary layer contributions in the TTL) are to the lifetime assumptions.

The following text has been added as a new subsection at the end of section 2.

"**2.3.3 The effect of assuming constant lifetimes**

The lifetimes of the halocarbons are not the same in the boundary layer and the TTL (Carpenter et al, 2014). The assumption of constant lifetime in a 12 day trajectory is evaluated by calculating the difference between idealised trajectories which had 2, 4, 6, 8, and 10 days in the boundary layer and 10, 8, 6, 4, and 2 days in the upper troposphere. Lifetimes for the boundary layer and for the upper troposphere for each gas were taken from Carpenter et al. (2014). (Lifetimes for higher altitudes are not available therein). The difference found between the two extreme cases are 6% ($CHBr_3$), 3% (CH_2Br_2) and 25% (CH_3I). The assumption is thus valid for the two brominated species.

This assumption is more robust than it might seem at first glance. The boundary layer fraction is calculated using 12 day trajectories in which there is little loss of CH_2Br_2 whether a lifetime of 94 or 150 days is taken. The most important factor in determining the amount lofted into the TTL is thus the original mixing ratio which is only slightly modulated by the chemical loss in 12 days. The longer lifetime is absorbed implicitly taken into account in the background contribution. The same arguments apply for $CHBr_3$, though the effect is a bit larger. The largest difference is seen for CH_3I. However, the difference matters much less for CH_3I because only 4-5% remains after the full 12 days which is much smaller than the uncertainties in this analysis so that much shorter trajectories are used to validate the new convection scheme."

(2) The presentation of the manuscript could be improved in several places. Specific suggestions are given below. Additionally, throughout the manuscript the authors should consider whether the citations given are the most appropriate to the points made in the text. An example is on Line 50 where the point is that

VSLS are emitted from the ocean and have natural sources. Given that, citations to modelling work looking at impacts of iodine/bromine chemistry (Solomon, Vogt, Salawitch, Saiz-Lopez) seem somewhat out of place. More appropriate and recent references would be, for example:

Hepach, H., et al. Biogenic halocarbons from the Peruvian upwelling region as tropospheric halogen source, Atmos. Chem. Phys., 16, 12219-12237, 2016.

Hepach, H., et al. Halocarbon emissions and sources in the equatorial Atlantic Cold Tongue, Biogeosciences, 12, 6369-6387, 2015.

Yang, G. et al. Spatio-temporal variations of sea surface halocarbon concentrations and fluxes from southern Yellow Sea, Biogeochemistry, 37 121(2), 369-388, 2014.

We have also read through it carefully and tried to improve the clarity. The point about the referencing is taken and we have added these and some other, more relevant references to the manuscript with that in mind.

Discussion paper

I suggest the authors thoroughly proof the paper for similar instances and areas where readability could be improved.

We have carefully read through the papers with a view to making it clearer to the reader.

Specific comments:

Line 38: The issue of "continued depletion in the lower stratosphere" is debatable. Mid-latitude and tropical ozone in this region is strongly influenced by transport and much of the apparent downward trend reported by Ball et al. appears to be have been reversed in 2017, as shown by Chipperfield et al. (2018). I would encourage the authors to amend this sentence to a more precise one. Chipperfield, M. P., Dhomse, S., Hossaini, R., Feng, W., Santee, M. L., Weber, M., et al. (2018). On the cause of recent variations in lower stratospheric ozone. Geophysical Research Letters, 45, 5718–5726. https://doi.org/10.1029/2018GL078071

We have added the reference to Chipperfield et al (2018). However we note the recent publication of Ball et al (2019) in ACPD and think the jury is still out. We have changed 'depletion' to 'possible reduction' due to the likelihood of its origin as being dynamic.

Line 47: on first appearance spell out the name of these compounds: i.e. methyl iodide (CH3I), bromoform (CHBr3) and dibromoethane (CH2Br2).

Names of these compounds have been spelled out.

Line 52: Is there a reason why specifically 12 days is chosen? In the Discussion (line 461), it is noted that longer periods are tested but the details are very vague. I would state earlier on in the manuscript that sensitivity tests were performed and be more quantitative on what was found.

See above

Line 82: "east" — "East"

This has been corrected.

In Section 2.1 it would be useful to indicate the altitude limits of the various aircrafts. Related to this, it would help the reader to know how the TTL is being defined up front.

Agreed. Altitude limits of the various aircraft have been added:

CAST BAe-164 0-8 km

Gulfstream V	1-14 km
Global Hawk	13-19 km

-Line 140: The citation to Jones et al. should probably appear directly after NAME.

Agreed – this citation appears directly after NAME.

Line 215: Should “Research Flights” have capital letters?

We have switched to lower case, except in section and caption titles.

Line 217: “very short lived brominated substances” could be deleted

Agreed. These words have been deleted.

Line 222: Starting a sentence with this number is a bit odd. Consider rewording or spelling out the number.

Agreed. This number has been spelled out.

Line 223: “TTL” — “the TTL”

Agreed.

Line 248: “western and central” — “Western and Central”

Agreed.

Line 387: define MJO

Agreed, the MJO has now been defined as Madden-Julian Oscillation.

Figure 1 caption: I recommend reworking as brackets within brackets looks odd here.

Brackets have now been removed and replaced with hyphens.

Figure 2 caption: What are the black symbols? Should also indicate if box and whiskers are the same as Figure 1.

Agreed, black symbols are the same as Figure 1 and represent measurements.

Figure 4: A reduced x-axis scale for each species would improve readability of the data.

We have used this scale to be consistent and for easier comparison of Figure 4 with Figures 1 and 6. We fully understand the suggestion and the reviewer’s intention to improve readability of the data by reducing x-axis scale but we would prefer to keep it unchanged to help readers compare the data between multiple figures. We are happy to accept the editor’s judgement on this.