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Interactive comment on "Variability and long-term changes of brominated VSLS at the tropical tropopause" *by* Susann Tegtmeier et al.

Anonymous Referee #2

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The manuscript presents a study of the transport of CHBr3 and CH2Br2, concentrating on how this might have changed over the past decades. They use a now widely-used static global sea-surface concentration field for these species, then calculate monthly fluxes based using offline meteorology. This is run through a Lagrangian model using emissions on a monthly basis. This is an area of study that is of interest to a wide audience and is appropriate in scope for ACP.

I can understand that the approach described here could yield information about the source gas delivered to the TTL/UTLS, however, I am not convinced that it is an appropriate methodology for understanding product gases. A largely expanded discussion of uncertainty is needed, as well as comparisons with product gases or referencing of this work elsewhere. The comparisons with inorganic bromine constraints could

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Discussion paper

provide evidence to the argument that this methodology can give valid insights into product gas delivery to the stratosphere. Clearer and more detailed explanations are needed for this approach, as these kinds of studies have more often used models with online chemistry with the online process (e.g. photolysis, deposition, heterogeneous chemistry) and are run at higher temporal resolution.

The approach to comparisons with the aircraft needs to be improved, both in presentation and approach. Comparisons of the entire observational period or simply a daily mean are insufficient. Comparisons with aircraft data have been done to a higher standard in the literature for many years. Comparisons should at least be made for the same location and time. Why was not a higher model timestamp chosen? The comments that the high concentrations are transient and not captured by models doesn't seen particularly novel, and also likely to be highly sensitive to this.

A more general point is that there must be a point at which the limitations from having a single static ocean-surface concentration dataset becomes a limiting factor in what science can be done. Are seasonally resolved predictions really possible without some exploration of the effect of seasonality in the initial concentrations? An opportunity seems to have been missed here to study the impact of perturbing this source of uncertainty.

The language used throughout the manuscript could often be improved. Standard manuscript preparation rules are not followed (e.g. abstract starting with some context for the work). The text requires updating and proofreading for grammar and clarity of language.

I cannot recommend publication of this manuscript in ACP without substantial updates as highlighted above and detailed below.

Specific comments

~Pg. 1 - Please include the word "past" or "historical" in the title and relevant points in the text. This means that the readers will more easily understand how the paper's variability fits with other recent work in the literature, including from the same authors (e.g. Ziska et al 2017).

∼Pg. 2 Ln 3 - Please start the abstract with context and motivated the work, rather than straight away stating what has been done here.

∼Pg. 2 Ln 4 - What is meant here by "available observational datasets"? Do the authors mean all observational datasets have been used, if not which ones and why?

∼Pg. 3 Ln 21 - Why is the word "currently" used hear when papers from 2006/2011 are cited. The authors are correct to point out that the heterogeneous chemistry and cycling of bromine has seen notable study recent. Please update this section based on recent literature (e.g. Fernandez et al 2014, Schmidt et al 2016 etc etc)

∼Pg. 5 Lns 16 - As before. Please clarify what is meant by "available observational data sets".

∼Pg.5 Lns 17 - The use of the terms "high-resolution" to describe 1x1 modelling is odd. This is not enough to really resolve the coastal regions that huge gradients in emissions of the species studied here are seen. Global models are now regularly being run at higher resolutions, such as at horizontal resolution 12x12 km (e.g. Hu et al 2018).

∼Pg. 6 Lns 3-4 - please include the word "static" to ensure the reader is aware that no temporal information is present in the concentrations. This is mentioned later in this section, but should be said at the beginning too.

∼Pg. 6 Lns 12-13 - The gap-filled dataset is therefore 6 years old? Have any measurements been made since that point? Specifically, in locations where effectively no observations are present (e.g. Indian ocean). Would these observations decrease uncertainties within the dataset? What are the uncertainties within the dataset for these regions? Would this mean there is enough data to get a seasonally-resolved dataset?

∼Pg. 8 Ln 10-12. Please add a brief discussion of the lifetimes of these species to the introduction. Also state here why the different run periods were used.

~Pg. 8 Ln 16. Why is the model run period outside the observational constraint of the emissions (stated as 1989-2011 in the text)? This should be stated in the text.

~Pg. 9 Lns 6-8 - What about in FLEXPART? Does this reproduce what is seen in TOMCAT? Even with the differences in timestep and online processing?

~Pg. 9 Lns 9-10 - What "photochemical" loss? Do the authors mean the prescribed loss terms they have used? What about loss through oxidation (e.g. OH)?

~Pg. 9 Lns 11-13 - This approach to Bry cycling and loss is much simpler than what is employed in the state of the art chemical transport models. What sensitivity studies have been performed to see the sensitivity to this? This seems likely to cause large differences in the product gases delivered to the stratosphere.

~Pg. 9 Lns 17-23 (I) - These authors have not convinced the reader that the partial use of offline partitioning is appropriate. The lifetimes of some of these inorganic species are of the order of minutes and partitioning dramatically changes from day to night. Why too are partitioning calculations being used from 10-15 years ago, have the values predicted by the field not changed? What are the differences between the simulations of inorganic bromine in these simulations and those (even in the same model) in more recent work (e.g. in CAM-Chem, GEOS-Chem, and TOMCAT). Either strong evidence that the answer is insensitive to this simplification needs to be added, or the discussion of product gases should be removed.

~Pg. 9 Lns 17-23 (II) - Please give a statement here about how the model has been evaluated against the observations for inorganic bromine (like a similar statement for the VSLS on Pg. 8 Ln 30-32). Multiple recent studies have reported these observations (e.g. Koenig et al 2017)

~Pg. 9 Lns 17-23 (III) - More explanation needs to be given of how the mixture of the online and offline process are considered here? There is a prescribed lifetime, does this just include photolysis or oxidation too? But it doesn't include wet deposition? Or are depositional losses being double-counted here?

∼Pg. 9 Lns 25-36 - Why not diagnosis the tropopause from the ECMWF met fields? This seems like an unnecessary simplification.

∼Pg. 10 Lns 3-5 The authors are right to point out this large uncertainty on their prediction of product bromine. However, a full discussion of this is essentially missing from the manuscript and must be added.

∼Pg. 17 Lns 1-11 What other model-observation comparisons have been done in the literature. Have these failed or succeed in capturing the observations? Please add this information to give context for the work.

∼Figure 2 - Why do units of concentration presented in Fig 2d in have a time dimension?

∼Figure 3 - Please explain the noisy pattern seen in the model values underlayed.

∼Figure 4 - This figure does not clearly convey information. Why are whole campaigns solely represented as stars? Why not show some level of variability seen by both the modelling and observations? It seems like a simple box and whisker comparison would convey more information here. Why two are the multiple horizontal lines, is this a split y axis? If so please show this with an axis break symbol.

∼Figure 7 - Please show uncertainties or ranges for the observations from ATTREX on the leftmost and rightmost subplots. Also, The paradigm for plotting in atmospheric science is usually that observations are shown in black and model. Please update this plot and anywhere else in the manuscript to avoid confusing readers.

∼Figure 8. (e) Please explain discontinuity seen at latitudes of -5 degrees N and 5 degrees N in the indian ocean. These do not seem physically plausible.

∼Pg. 37 Ln 1 - Are only the bromoform emissions archived? What about the dibromomethane, where is this data archived? Technical comments

~Pg. 2 Ln 22 - expand “Br/dec” to include “decade”.

~Pg. 5 Ln 5 - Typo - “meteorological”

~Pg. 5 - Ln 23-24 - Sentence does not scan well. Please rephrase. “The question if such hotspots are mainly driven by oceanic or by atmospheric processes will be answered based on the Lagrangian simulations.”

~Pg. 8 Lns 4-7 - Please update the sentence as below for readability. The sentence structure here and elsewhere could be more concise.

From “with the Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005) 7 Version 9.2 beta.”

To: From “with the FLEXPART Lagrangian particle dispersion model (Version 9.2 beta; Stohl et al., 2005).”

~Pg. 11 Lns 25-27 - “In the following,” is verbose and should be removed.

~Pg. 35 Ln 18 - Add spaces between numbers and units here and elsewhere in the manuscript. Please refer to NIST guidance on this (https://physics.nist.gov/cuu/Units/checklist.html)

~Pg. 11 Ln 25 - Both analyze and analyse are used in multiple places in the text. Please choose to use either the American or British version.

Refs.

Fernandez, R. P., Salawitch, R. J., Kinnison, D. E., Lamarque, J.-F., and Saiz-Lopez, A.: Bromine partitioning in the tropical tropopause layer: implications for stratospheric injection, Atmos. Chem. Phys., 14, 13391-13410, https://doi.org/10.5194/acp-14-13391-2014, 2014

Hu, L., Keller, C. A., Long, M. S., Sherwen, T., Auer, B., Da Silva, A., Nielsen, J. E., Pawson, S., Thompson, M. A., Trayanov, A. L., Travis, K. R., Grange, S. K., Evans, M. J., and Jacob, D. J.: Global simulation of tropospheric chemistry at 12.5 km resolution: performance and evaluation of the GEOS-Chem chemical module (v10-1) within the NASA GEOS Earth system model (GEOS-5 ESM), Geosci. Model Dev., 11, 4603-4620, https://doi.org/10.5194/gmd-11-4603-2018, 2018.

Koenig, T. K. et al. : BrO and inferred Bry profiles over the western Pacific: relevance of inorganic bromine sources and a Bry minimum in the aged tropical tropopause layer, Atmos. Chem. Phys., 17, 15245-15270, https://doi.org/10.5194/acp-17-15245-2017, 2017.

Schmidt, J. A., et al. (2016), Modeling the observed tropospheric BrO background: Importance of multiphase chemistry and implications for ozone, OH, and mercury, J. Geophys. Res. Atmos., 121, doi:10.1002/2015JD024229.

Ziska, Franziska, et al. "Future emissions of marine halogenated very-short lived substances under climate change." Journal of Atmospheric Chemistry 74.2 (2017): 245-260.

END OF REVIEW

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