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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Bromine from short–lived source gases in the Northern Hemisphere UTLS" *by* Timo Keber et al.

Anonymous Referee #1

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Review ACP-2019-796

Bromine from short-lived source gases in the Northern Hemisphere UTLS

1 general comments

`evaluating the overall quality of the discussion paper`

The paper presents novel observational data of VSLS from several aircraft campaigns and compares them with modeling results. It extends on and confirms previous find-

ings. Hence, the title does reflect the contents of the paper.
Some scientific methods and assumptions need more thorough outline and proper discussion.
The results are sufficient to support the interpretations and conclusions, but not all results can be repeated based on the information given, e.g. the derivation of the tropopause for the campaigns labeled as "private conversation".
The authors give proper credit to related work and indicate their own contribution, but they could make it more clear from the start, what the current consensus regarding the emission scenarios is.
The abstract provides a complete summary, but it does not deliver regarding the WHY unit, e.g. the context and importance of the measurement. It gives the impression of a paper focused on observational data, whereas 2/3 of the figures and text are related to model evaluation. If the point of the paper is to evaluate existing emission scenarios through a new set of data, this should be made more clear. The overall presentation is well structured and clear.
The language is fluent. The authors should be more cautious with the usage of the term "significant". Some parts of the paper need editing. In particular, the manuscript does not follow the ACP guidelines in several points:

- Subsections should be consecutively numbered.
- Figure captions, figures, and tables:
  - Usage of full campaign names renders the captions imprecise and utterly unpleasant to read.
  - Poor choice of colors (red, green, blue, black, grey) within the line plots (vector graphics!) and the tick labels' font size make the figures hard to read (Fig. 7, Figs. 13-15).
  - The usage of "[ ]" around units in plots is depreciated (→ https://www.bipm.org/en/publications/si-brochure/section5-3.html)

- Some white spaces seem odd.
- Equations are not properly set, e.g. usage of "*" as indicator for multiplication.

The number and quality of references are appropriate.

2 specific comments

`individual scientific questions/issues`

- P4L33: *"It is clearly visible that the halocarbons correlate [...]"* Can you quantify this?
- P5L40-P6L1: *"Only bins which contain at least five data points [...]"* What criterion led to this choice? (See comment to Fig. 4 below.)
- P6L21-22: *"[. . .] the variability averaged over the four lowest stratospheric bins was always lower when using $\Delta\theta$ as a coordinate."* Can you elaborate on this? Why does the variability in the four lowermost stratospheric bins change in response to the transformation of coordinates $\Delta\theta$ (relative to the tropopause)? Am I right to assume that this is due to bins with mixed tropospheric/stratospheric data? If that is the case, "four lowest stratospheric bins" is misleading. Please elaborate on this.
- P7L5-7: *"The data have been binned in 5° latitude and 5 K intervals of potential temperature. As expected, the distributions closely follow the tropopause (indicated by the dashed line), with values decreasing with distance to the tropopause and also with increasing equivalent latitude."* In the previous section the authors

used *"[o]nly bins which contain at least five data points [...]"* in a much coarser binning. From the sampling frequency, one ought to assume that the shown averaged VSLS concentrations per bin are based on similarly sized numbers of entries, but this neither emerges from the text nor the figures (Fig. 5, Figs. 9-12). Furthermore, earlier in the manuscript the authors find that $\Delta\theta$ is the coordinate of choice for this study, but they don't use it in these figures. If it was possible to show the data in relative coordinates, this would strengthen their point (*"closely follow the tropopause"*). The authors should elaborate on this.

- P7L7-9: *"The distributions observed during the WISE and the TACTS campaigns show significant amounts of CH_2Br_2 [...]"* What purpose does "significant" serve in this context? Elaborate on the actual significance or drop the word.
- P7L10-16: *"[. . .] the most stratospheric air [...]"* and *"[. . .] very high mean age of air [...]"* If the authors' point is to state that the amount of VSLS in the stratosphere is a function of its residence time there, they should make this more clear.
- P7L28-29: *"[. . .] are significantly larger [...]"* Can you quantify this? Else drop the term "significantly".
- P7L36-38: *"[. . .] significantly slower [...]"* Same as above.
- P8L10-12: *"[. . .] has been extended from the ESCiMo simulations to cover our campaign time period (see Section 2)."* The authors give little detail about the extent of this extension. In Section 2, this "extension" of the specified dynamics simulation from the original ESCiMO simulations is not even mentioned at all. The authors refer to Jöckel et al. (2016) for the description of the set-up used. However, Jöckel et al. (2016) give at least 4 simulation set-ups (e.g. RC1SD-base-07/10/10a, RC2-base-04) on which this extension might be based. Presuming this extension includes the time span 2014-2017, it is not clear which prescribed tracer emissions were used. The authors need to provide more details.

- P8L25-28: *"As no direct tropopause information was available for the TOMCAT output [...]"* Refering to Section 2 the authors state: *"Local tropopause information for the flights with HALO have been derived from ERA-interim data [. . .]"* Could the tropopause information from ERA interim, with which the CTM was driven, be derived?

- P8L29: *"[. . .] EMAC tropopause and the climatological tropopause differed by less than 3 K[...]"* How is the EMAC tropopause defined? How does it differ from the WMO/PV definition? In principle, they are comparing a "modeled" tropopause in EMAC, which has been nudged to ERA interim, with a tropopause climatology directly derived from ERA interim. They do not discuss that fact. What is the point here? Temporal and spatial stability of the tropopause in the time frame of interest?

- P9L10-18: The authors are referring a sensitivity study conducted within the EMAC framework's quasi chemistry-transport model (QCTM) mode (Graf 2017). Four different emission scenarios for VSLS are compared therein. They conclude their qualitative comparison of Figs. 7, 8 with the conclusion: *"It is therefore clear that the observed differences are not primarily caused by the model but rather by the emission scenarios."* With respect to the shape of the vertical VSLS concentration profiles, this may actually be true. But this statement does not hold if one takes a closer look at, e.g. the VSLS concentrations at the tropopause between EMAC and TOMCAT. On the back of an envelop – normalize the EMAC QCTM sensitivity studies (Fig. 8) with respect to the Warwick scenario and compare, e.g. Ziska scenarios, with TOMCAT in Figure 7. One finds that TOMCAT mixing ratios of CH_2Br_2 at the tropopause seem to be about 0.3 ppt higher than in EMAC. The spread between the scenarios is roughly 0.5 ppt (0.2 ppt if the Warwick scenario is excluded). Hence, there seems to be a substantial difference between the models, too.

- Section 5: The authors describe a "simplified approach" to estimate total and inorganic bromine from VSLS in the extratropical stratosphere. They assume a linear mixing of two air masses, wherein the fraction of bromine with tropical origin increases linearly with increasing $\Delta\theta$. The bromine mixing ratios are only evaluated once, at the tropical (TTP) and extratropical tropopause (ExTTP). This ansatz completely neglects the actual transport (horizontal as well as vertical) and therefore the further photochemical transformation (depletion) which VSLS undergo. Based on kinetics, one would expect the concentration of any of the VSLS (let's call it [A] for 'any') at the tropopause ($[A](0,0)$) to follow a powerlaw:

$$[A](t,\theta) = [A](0,0) \cdot e^{-k\cdot(t(\theta)-t_0)}, \tag{1}$$

  with e.g., $k \propto J_a + k_A(T)[OH]$. If we look at [A] at $\Delta\theta$ and a time t' and assume the same linear superposition of air masses as the authors:

$$[A](t',\delta\theta) = (f^{\mathsf{ex-trop}} - 1)[A]_{\mathsf{TTP}} \cdot e^{-k\cdot\Delta t_a(\Delta\theta)} + (f^{\mathsf{ex-trop}})[A]_{\mathsf{ExTTP}} \cdot e^{-k\cdot\Delta t_b(\Delta\theta)} \tag{2}$$

  Although the authors state *"[. . .] for models it is necessary to have a realistic representation not only of chemistry but also of transport in the lowermost stratosphere."*, this is not discussed, e.g. in a critical evaluation of the approach itself. Consulting Figure 4, the linear approach seems to be justified for $[CH_2Br_2]$, whereas $[CHBr_3]$ clearly follows a powerlaw. This is not discussed properly in the manuscript. The authors should reevaluate their approach and change it (if possible) or at least discuss it more thoroughly.

- Figure 4: It is not clear whether the shown error bars refer to the standard deviation (σ) in each bin or to its standard error ($s = \sigma/\sqrt{(N)}$). Latter is a more reasonable choice. In any case, displaying data in such way does only make sense as long as the distribution in each bin is Gaussian. Otherwise, a violin plot may be more valid. Have the authors checked their distributions?

- Figure 7: Caption L11: *"[...] extremely high values [...]"* What is "extreme" in this context? Which exclusion criterion has been used? The manuscript doesn't provide any further information on this. Please elaborate on this matter in the appropriate section.

3 technical corrections

purely technical corrections

3.1 General

Typesetting of the name "Ordóñez" in citations is not coherent and incorrect at times.

3.2 Specific

- P1L22: *"Distributions [. . .] below the tropopause shows [...]"* → "show"
- P1L26: *"A scenario which has emissions most strongly concentrated to low latitudes [...]"*. This sentence needs rephrasing – maybe: "A scenario with emissions mainly confined to low latitudes [...]"
- P3L8-10: *"Two pathways for input of halogens from short-lived gases are discussed: Source Gas Injection (SGI),where the halogen is transported to the stratosphere in the form of the source gases; and Product Gas Injection (PGI), where photochemical breakdown products of source gases are transported into the stratosphere, usually in inorganic form (i.e. Bry)."* This sentence is not concise. Please rephrase.

- P3L11-12: *"While halogens transported into the stratosphere due to PGI are usually directly in a form available for catalytic ozone depletion reactions, halogens from source gases must first be released in the stratosphere photochemically."* You may rephrase this along the lines: "Halogenes from product gases are readily available for catalytic ozone depletion reaction. Source gases have to undergo a photochemical transformation first."
- P3L23: *"The main source of brominated VSLS is believed to be[...]"* Although orally excepted, one should refrain from the usage of "believe" in a scientific context. Please rephrase. You may use, e.g., "most likely" or "observations indicate".
- P3L41-43 and P4L1-3: *"These observations are compared [...]"* and *"[. . .] these observations are [. . .] presented [. . .] and compared [...]"*. These sentences are almost identical. You may merge them into the latter.
- P4L7-11: *"An isothermal channel uses [...]"* and *"The second channel [. . .] uses [...]"* You may rephrase these in passive voice.
- P4L31,33: *"mean age"*. Supposedly "mean age of air" as used later on in the manuscript.

  Section 2: The authors only mention the spatial resolution of their models. It might be worth to mention the temporal resolution of the model output, too.
- P6L28: *"As in previous work [...]"* → "works"
- P6L28-33: First of all, most of this is a repetition of the text written in the beginning of section 3 and can be dropped. *"As in previous work [...]"* and *"However, we propose a somewhat different approach [...]"* These two sentences are slightly contradicting. They can and should be merged along the lines: "We slightly diverge from approaches in previous works [...]"

- P6L36-38: *"In order to ensure that this tropopause value is representative [...]"* This sentence is too long and might be grammatically incorrect (*"[...] when we have observations [...]"*). Please rephrase.
- P7L7-9: *"[...] which has a rather long lifetime in the cold upper troposphere and lower stratosphere (Hossaini et al., 2010) even quite deep into the stratosphere."* This sentence is quite unclear and needs rephrasing.
- P7L9-10: *"[...] is strongly depleted [...]"* Can you quantify this statement?
- P7L10-12: *"[...] flight levels [...]"* You may refer to "flight altitudes" instead.
- P7L36-38: *"This is most likely [...]"* The authors may rephrase this sentence into two. The natural breakpoint would be *"[...] with latitude. As [...]"*.
- P8L6-7: *"[...] are based on the emission scenario by Warwick et al (2006) [...] was run with different emission scenarios (Ordoñez et al., 2012;Ziska et al., 2013;Liang et al., 2010)."* The authors should mention which or the 8 scenarios by Warwick et al (2006) and which of the three by Liang et al. (2010) they have used.
- P8L36: *"overestaimtion"* → overestimation
- P9L4-6: *"Because of the different chemical lifetimes [...] above 20 K above the tropopause [...]"* This sentence might need some rephrasing. The authors may use $\Delta\theta$>20 K.
- P9L10-12: *"In Order to [...]"* This sentence is too long and not concise. Please rephrase.
- P9L12-13: *"Note that these simulations [...]"* You may drop "Note that"

- P9L23-24: *"Again, we use equivalent latitude*[...]"* This has been stated several times by now. As the authors indicated by using *"[a]gain [...]"*. They should consider dropping the whole sentence or the "again" therein.
- P9L39: *"The direct comparison of the distributions between the different model data sets is also interesting."* This statement is redundant. Please rephrase, e.g. "We will have a look at [...]"
- P11L8: *"In this Section [...]"* Add a comma after "section".
- P11L33: The indent of the equation number is incorrect. The authors may reduce the equation to $\mathrm{Br}_{\mathrm{tot}}(\Delta\theta) = \mathrm{Br}_y(\Delta\theta) + \mathrm{Br}_{\mathrm{org}}(\Delta\theta)$
- P12L20: *"Figure 14 compares [...]"* There is a grammatically issue here. A figure cannot compare anything. Please rephrase using passive voice.
- P13L18: *"have e a"* → Remove the "e" in between.
- P13L30: *"[...] a large dataset [...]"* Can you quantify this?
- P13L36: *"[...] which is line [...]"* → "which is in line"
- P13L40: *"in high latitudes"* → "at high latitudes"
- P14L3: *"[...] with large differences produced by the different emissions."* It may be better to use "caused" in this sentence.
- P14L3-5: *"Overall, for* $\mathrm{CH_2Br_2}$ *[...]"* Please consider rephrasing this sentence.
- Figure 1: The figure is too wide. Would it be possible to color code the different flights temporally?
- Figure 2: Is is possible to highlight the flight path displayed in Figure 3?

- Figure 4: Caption L8: *"dotted"* → "dashed"
- Figure 7: Referred to choice of colors under "general". Caption L10: *"bene"* → "been"
- Figure 14: Please include the information given in L1 of the caption also in the legend of the plots.
- Figure 15: The x-axis label of the lowermost panel seems to be incorrect (compared to the others in the figure) → *"Br total and Bry from VSLS"*. Caption L1: *"[. . .] at $\Delta\theta$ of 40 K [...]"* → "at $\Delta\theta = 40K$"