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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.

Rafael Pedro Fernandez (Referee)

rpfernandez@conicet.gov.ar

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The paper presents a complete set of carbon-bonded VSLS source gases (SGs) measurements performed in the Northern Hemisphere mid-latitudes using a GC-MS instrument on board an aircraft, as well as a comprehensive comparison of the observations with a complete set of model simulations oriented to evaluate the contribution of tropical and extra-tropical injection of VSLS to the lowermost stratosphere. The main results of the work are: i) the troposphere-to-lowermost-stratosphere transport of VSLS SGs through the extra-tropical tropopause is larger than that occurring within the tropical tropopause; ii) the contribution of both tropical and extra-tropical VSLS injection must be considered in order to reproduce the VSLS abundance within the mid-latitudes lower stratosphere below 400 K; and iii) the models and inventories used in this work show

certain limitations in reproducing the VSLS reactive transport and estimate the release of inorganic bromine (Bry) in the NH lower stratosphere. The paper also includes a seasonal, latitudinal and vertical analysis of VSLS abundance in the UTLS.

I found the paper very interesting and very well organized, presenting results in a clear and comprehensive format, and including interesting and constructive discussions. It is worth noting that even when the altitude/latitude-dependent observations itself would be worthwhile to be published, the authors have decided to go forward and present a comprehensive model-observation inter-comparison, which contributes to improves the general understanding of the reactive-transport efficiency of VSLS species within the UTLS. In particular, I found very descriptive and intuitive the vertical coordinate system ($\Delta\theta$ and θ*) they used to represent all results relative to the altitude of the tropopause, which allows a consistent description of the vertical and latitudinal decay of VSLS once they are injected to the stratosphere. At the very end of the paper, a simplified approach (eq. 4) is used to estimate the total amount of inorganic bromine (Bry) released by VSLS within the mid-latitude lower stratosphere, highlighting the major importance of properly reproducing these Bry levels in model simulations oriented to determine the ozone impact of VSLS.

Having said this, I believe that the manuscript posses a handful set of specific issues and many technical details (including figures and tables captions) that must be corrected before final publication.

Major Concerns:

1. I would like to start mentioning that most of the "important questions" that came to my mind while I was reading the manuscript had already been responded (i.e., as I moved forward with the lecture and reached Section 5). Even when this should be taken as a mainly positive comment, it also implies that some of the analysis/discussions given at the end could be (at least partially) shifted to earlier sections, to help the reading and support the analysis. For example: Sections 3 is concentrated on carbon-bonded

VSLS mixing ratios close and above the tropopause in the lower stratosphere, but there is only a brief mention and sideway comparison with the SGI values compiled in the last WMO report (P7,L27). The reader needs to wait until Table 5 is presented (Section 5, P12) to reach a complete discussion and comparison with WMO values, and it is only at this point that the importance of the model-observation inter-comparison (presented in Section 4) becomes evident. In doing so, note that Table 5, which provides values for VSLS SGs within the tropics and extra-tropics, is introduced in the only section of the paper focused on PGs (Section 5). 2. Observations from TACTS and WISE campaigns have been merged into a unique dataset (WISE_TACTS) because they were performed during the same seasons. Even though I found this procedure correct, I wonder if the authors have analyzed this data separately to evaluate if there is at least any glimpse of VSLS SGs trend within the NH-UTLS (there are $\sim$5 years between both campaigns). The authors declare they combined both dataset based on "general" observational evidence (P3,L29), but I think they should justify this procedure by evaluating specifically their unique and novel dataset (P4,L29).

3. Authors should be really careful and consistent when using the wording "total bromine". Until Section 5 is reached, only carbon-bonded (i.e., organic) bromine is considered, and total bromine is referred as the sum of CH2Br2 + CHBr3 + minor_VSLS (P6,L2; P8,L15). But later on Sections 5 and 6, "total bromine" points out to the sum of Br_org + Br_inorg (P11, Eq. 1). Please, be consistent and refer to "total organic bromine" and/or "total bromine" whenever appropriate.

4. Although the vertical profiles for CH2Br2 and CHBr3 are analyzed in detail, there are no mentions regarding the "error bars" presented in the figures (e.g., P6,L20; Fig. 4 caption). Do the error bars correspond to 1-sigma or 2-sigma? Besides this, the authors should explain why the vertical error bars for $\Delta\theta$ are not the same for the different bins, as well as why the error bars for CHBr3 are larger than for CH2Br2? Is this only due to the shorter lifetime of CHBr3, which shows quite different vertical profiles depending on the exact latitude within the 40-60° bin (as observed in the latitude-altitude

cross-sections), or could this also be attributed to differences in their regional source strengths? Note that the variability is only considered in (P7,L34; Fig. 6) when addressing how the VSLS tropopause abundance changes with latitude, but I think that a more complete comparison of this latitudinal and vertical variation within the 20-40°N bin should be provided (at least the two major VSLS).

5. P8,L18: "The contribution from these mixed bromochlorocarbons to total VSLS bromine are typically on the order of 20%, while about 80% of total VSLS bromine in the upper troposphere and lower stratosphere is due to CH2Br2 and CHBr3". $\rightarrow$ Are this percentages computed using WISE_TACTS and PSG data? Have you compared this findings with other studies (i.e., Fernandez et al., 2014)? Have you thought about presenting combined results of your observations of the sum of these minor_VSLS into a figure or table? (currently only results for the sum of CH2Br2 + CHBr3 + minor_VSLS are given)? As minor_VSLS posses in some cases lifetimes larger than CH2Br2, this information could be useful for future studies. The importance of the minor VSL contribution becomes also evident in Section 5 (P13,L11) when the overall contribution from longer-lived VSLS to Bry is discussed.

Specific Comments:

1. Title and Abstract: Shouldn't the title be more specific on the extra-tropical (or the "tropical vs. extra-tropical") contribution of VSLS bromine to the UTLS?? In addition, the abstract has an excessive focus on the results obtained with the different models and emissions scenarios (including specific statements for some of the scenarios giving the best and worst agreement). I would expect the abstract to focus on the contribution of tropical vs. extra-tropical contribution to SGI, and in any case to provide a rough estimate of the relative contribution of each of these two pathways to the overall organic and inorganic lower stratospheric bromine (and providing only a general mention to the similarities and discrepancies between models and observations and its dependence on latitude and season).

2. Section 3 (P5): The first paragraph of section 3 describes the spatial and vertical coordinates used for representing measured and modeled data. In my opinion, the selection of θ, $\Delta\theta$ and θ* variables really improves the analysis and interpretation of the results. However, I believe the initial description of how these variables are computed is not clear enough, and the reader needs to go back and forth between figures and text to completely catch up the difference among them. For example: a) $\Delta\theta$ is mostly used for vertical profiles figures, whereas θ* is used for latitude-altitude cross-sections (which is not clearly mentioned in the text); b) there are at least 2 or 3 places where $\Delta\theta$ and θ* vertical coordinates are defined, and in some cases slight differences on the definitions are observed (P5,L31 and P6,L32). In particular specify if the vertical coordinate is computed above the "local" tropopause "for each latitude" and how is it added to the "climatological" tropopause.

3. P11,L2-5: "Most importantly, the overall levels, especially in the low latitudes, are much higher than our observations and also much higher than the tropical observations compiled in the WMO report (Engel and Rigby, 2018). This will result in too much VSLS bromine being simulated in the stratosphere, and therefore also in a misrepresentation of the input to the lowermost stratosphere via the different pathways". → In addition to the general description focused on the 340-400 K range, I found interesting that poleward of 40° and below 320 K (see Fig. 9-10) there is a negative model bias for CH2Br2 mixing ratio exactly at the extra-tropical tropopause, while at the same time there is a positive model bias for CHBr3. This is not mentioned nor explained in the text. 4. P11,L28: The simplified approach considering fext-trop and ftrop" is very intuitive and helps to visualize the contribution from tropical vs extra-tropical bromine from VSLS, but it would be useful to provide at the end a conclusive sentence of which are the most probable fractional contributions from tropics and extra-tropics to the overall bromine in the UTLS. Certainly, concurrent Bry measurements would be required to close the whole bromine budget, but at least from Figure 15 ($\Delta\theta$ = 40 K) it seems that fext-trop values close to 20-40% produce the closest agreement between model and observations. However, on the analysis presented for Figure 14, fext-trop is 60% at

this height, isn't it? I suggest to expand the analysis and discussion of fext-trop and ftrop" (you only dedicate a few lines to this subject at the end of Section 5). Finally, why are not equivalent results for WISE_TACTS provided in Fig. 15? 5. P12,L28-29: "The larger Bry derived in the model calculations above 60 K is caused by the higher total bromine values from CH2Br2, which are caused by the higher CH2Br2 levels at the tropical tropopause in comparison to the observations." P12,L32-33: "In the lower part the discrepancy is more due to higher simulated CH2Br2 in the lowermost stratosphere than found in the observations" → This could "partially" be the reason, but it could also be due to using an improper fext-trop value at this specific vertical level and/or due to the simplified linear approach used. Please elaborate on this. 6. P14,L29: "we have shown that there will be significant differences in stratospheric Bry depending on the emission scenario, which can be as high as 2 ppt, corresponding to a difference of a factor 2 relative to observation-derived values". → Being this sentence included in the conclusions (and also mentioned in the original abstract), I suggest informing not only the largest (i.e., worst) difference, but also the minimum model-observation differences, as well as the range of model bias results for the models which show a better performance.

Technical Corrections:

P1,L17: "The instrument is extremely sensitive due to the use of chemical ionisation, allowing detection limits in the lower parts per quadrillion (10 -15) range". → Is this information of major importance to be included on the abstract? Consider also including the GS-MS acronym in the preceding sentence.

P1,L35: "Depending on the underlying emission scenario, differences of a factor 2 in reactive bromine derived from observations and model outputs are found for the lowermost stratosphere, based on source gas injection." → Consider rephrasing, and also mentioning the range of agreement of models (see comment above).

P3,L30: "A further future increase has been projected" → A future increase of VSLS

emissions has been suggested"

P3,L38: "In order to investigate the regional variability of bromine input into the lowermost stratosphere and the inorganic bromine loading of the extratropical lowermost stratosphere, we have performed a range of airborne measurement campaigns ..." → You explicitly mention "inorganic bromine" but not "organic bromine" in a sentence focused on the novel measurements dataset. Consider revising, as you've only measured carbon-bonded species, and only inferred inorganic bromine.

P3,L42;P4,L3: Please specify which are "the implications" you are pointing at.

P4,L5: Consider changing the subtitle to "Instrumentation and Observations"

P4,L22: Check for consistency between the year of the TACTS campaign between the text (2011) and table 2 (2012).

P4,L29: "covered a similar time period and latitude range" → you mean same seasons, consider rephrasing.

P5, L2: "ESCiMo (Earth System Chemistry ntegrated Modelling)" → Integrated

P5,L12: TOMCAT acronym already defined above (P5,L2).

P5,L7;P5,L13: If both EMAC and TOMCAT are driven by exactly the same ECMWF ERA-Interim reanalysis data, this should be mentioned explicitly. This will help to override additional uncertainties regarding differences between models. Also, although EMAC can be run as a CCM model, it should also be clear that for the current SD simulations, the model behaves like a CTM.

P5,L17: "Emitted VSLS (CHBr3, CH2Br2, CH2BrCl, CHBr2Cl and CHBrCl2) are destroyed by reaction with OH and photolysis in the model" → I assume this is also the case for the EMAC model described in the preceding paragraph. You should explicitly mention this to avoid confusion.

P5,L24: Although the paper reads perfectly well using the θ vertical coordinate, it

should be mentioned at least during the model description which are the equivalent altitude/pressure values for the tropical/extratropical tropopause θ levels used in this work.

P5,L36: "we have also binned the data in potential temperature in 10 K potential temperature intervals" → repetitive

P5,L38 and elsewhere: "relative to the mean tropopause observed during the campaigns" → in many places the authors make reference to "the campaigns" in a general meaning, when I understand they are pointing out to "each dataset" obtained during the campaign, and not the campaign itself.

P5, L38: "The results are presented ..." → consider rephrasing the whole sentence, as it is very difficult to understand. It should also be mentioned at least once that whenever you mention winter, spring or fall, you are always pointing out to "boreal" seasons. It is not necessary to repeat it all over the text, but I only found it mentioned properly once in the conclusions (P13, L39).

P6,L15: "again in line with their atmospheric lifetimes, which generally decrease with an increase in the bromine atomicity of the molecule". Atomicity should also be used in P7,L25.

P7,L10: "The shorter-lived CHBr3 is strongly depleted already about 20 K above the tropopause" → Based on Figure 5, this is only the case during PGS, but not for WISE_TACTS during the summer. Could this be related to stronger convective transport during the summer?Please explain.

P7,L20-22: "In order to ..." → there are many sentences that begins with this wording. Although I found it correct, please avoid it using more than once within a paragraph (i.e., here it is used in two consecutive sentences).

P7,L23: "Again, for the tropospheric data, standard latitude has been chosen, while equivalent latitude was used for all data with $\Delta\theta$ above zero" → This got me confused:

Figure 6 focus on extra-tropical tropopause values (averaged considering bins 10 K below the local tropopause). Wouldn't this correspond to negative $\Delta\theta$?

P7,L29: explicitly point at Tables 3 and 4.

P7,L30 and Figure 6: Could the "negative latitudinal gradient" observed for WISE_TACTS be somehow unrealistic/largely-biased because of using a small amount of measurements below the tropopause at larger latitudes? Or because of any type of cos(lat) averaging factor?

P7,L34: "derived around the tropopause" → wouldn't it be "below" (-10 K) the tropopause.

P8,L1-5: Sentence is too long. Please rephrase.

P8,L13: "Here we compare vertical profiles, geographical distributions and latitudinal gradients between our observations and the model results". → What do you mean by "geographical distributions"? 20°-40° bin?? If not appropriate, please remove.

P8,L23: "about 40 K" → "∼40 K"

P9,L4: "Using the Ziska et al. (2013) emission scenario, the overestimation of CH2Br2 and the underestimation of CHBr3 tend to cancel out, resulting in a reasonable agreement in total VSLS bromine. Because of the different chemical lifetimes of the two species, this results in a wrong vertical distribution of Bry with too high mixing ratios above 20 K above the tropopause in winter and a much steeper vertical gradient in late summer." → First, in the initial sentence it should also been explicitly mentioned that, in addition to the CH2Br2 overestimation and the CHBr3 underestimation, the contribution form "minor VSLS" which is also considered for the total "organic" bromine results is based on the Ordoñez inventory . . . adding an additional uncertainty to the different contributions that "cancel out" each other. Second, What do you mean by "vertical distribution of Bry" in this context?. Please be careful when pointing out to the inorganic or organic bromine in this section.

P9,L23: Fig. 9 to 12 instead of 9 and 10?

P10,L6: "much lower" → please be more specific

P10,L15: "Therefore, we compare the observed mole fractions of the brominated VSLS in the upper troposphere with those determined from the different model setups, in order to investigate if the models are able to represent the latitudinal gradient in upper tropospheric mole fractions." → please rephrase.

P10:L17: "or respectively modelled (EMAC)" → please rephrase (see comment on respectively below)

P10,L32-37: The text goes back and forth a couple of times between wintertime results and summertime results. It would be simpler to describe all results for one season before moving to the other.

P10,L36: Here and elsewhere . . . "extremely high" → what do you mean by extremely? Wouldn't just "larger" be enough?

P11,L12: "is expected to add more bromine on top of SGI"

P11,L14: "imply for the total bromine and inorganic bromine" → you mean total organic bromine or total (organic + norganic) bromine? Or both? Please see the general comment above.

P11,L18: Too many "and" within a single sentence. Please rephrase

P11,L23: "No studies on mass fractions are available for the campaigns discussed here, so we will rely on previous studies for these fractions." → I do not understand the rationale for including this sentence. Please make it clear or remove.

P11,L24: "order of magnitude difference" is normally used to point at scaling factors like 10, 100, 1000. Please rephrase.

P11,L41: Here and elsewhere. "at the tropical, respectively extratropical (40-60°N)

tropopause," → It is very unfamiliar for me the way the "respectively" sentences have been written throughout the text. I suggest replacing them by "at the tropical and extratropical (40-60°N) tropopause, respectively". See also P12,L19-L23.

P12,L6: "by averaging the model respective observations" → This sentence is senseless, in any case "the model results"

P12,L14: fext-trop decreases with altitude, not increases.

P14,L6: "with a downward revision" → do you mean shifted to the lower edge of the range of emissions?

P14,L26: "The bromine budget in the lower stratosphere will depend on the relative fraction of air from the tropical and extratropical tropopause. The relative contribution of extratropical air will decrease with altitude and should reach zero at about 400 K potential temperature.." → is it the future (will) tense appropriate here??. In any case, I believe the authors are pointing at a decrease with altitude and not latitude here (if not, please explain the idea and make it clear).

Tables and Figures:

Most of Figures and Table Captions include the "long-name" of each of the Campaigns instead of just providing their "short-name"/acronym (PSG=POLSTRACC+GW-CYCLE+SALSA, TACTS_WISE, HALO, ECMWF, etc.), which is not only simpler, but also more familiar to everyone. Using the short-name version will certainly improve the captions readability. Also, there is no need to define ppt each time you use it (only once at the beginning within the text is enough).

All Tables: Consider using a "one line title" at the top of each table, and then provide all specific information regarding the season, specific campaign, altitude/latitude range, etc. as footnotes on the table.

All multiple-panel Figures should indicate, in addition to the (letf, PSG) and (right,WISE_TACTS) information, that results are provided for (top, CH2Br2), (middle,

CHBr3) and (bottom, total organic bromine). (This example was based on Fig. 13, and should be adapted to the specific figure).

Table 3 and 4: What does "TP" stands for (tropopause)?. Why Table 3 provides values for TP + 30-40 K, while Table 4 only for TP + 40 K? Define what does the stdev. stands for and how is computed? Wouldn't it better to provide the stdev. value with a +/- sign (0,55 +/- 0,09) ppt within the mole fraction column?

Table 5: Have you used the annual mean or the seasonal mean for computing the tropical tropopause values?? (P12,L6). In case a seasonal mean has been used, please specify which months have been used for the model output. Replace bromide by bromine. Rephrase respectively. Explicitly indicate that the Br_ext-trop and Br_trop are for ($\Delta\theta = 0$). Indicate that ML stands for Mid-latitudes, which you called extra-tropics throughout the text. Define explicitly in a table footnote the $\Delta\theta$, latitude range and any other relevant information that has been used for computing the values presented in Table 5.

Figure 4: What are the horizontal and vertical bars? 1-2 sigma? Also, in the text it would be good to explain within the main text why for WISE_TACTS there are some points for which the mixing ratio as a function of $\Delta\theta$ can be computed ($\Delta\theta$ = 100), but not for the θ coordinate (θ = 420 K).

Figure 5: Consider introducing a dashed/dotted vertical line indicating the 40-60° boundaries (and any other important latitude) used for the extra-tropics vertical profile computation.

Figure 7: Consider reducing the length of the caption, and moving some of information at the end of the caption to the main text.

Figure 8: Specify output for this simulations is not in SD mode as for the other simulations (here and in the main text).

Figure 9-12: Consider including vertical dashed/dotted lines as suggested for Fig. 5.

Also . . . for the model (right) panels: Why there are some “empty/blank” boxes within tropical lower stratosphere above 400K? Is that because of the vertical model resolution and/or upper limit of the models?

Figure 13: Have you considered the idea of expanding the lower latitudinal edge of the figure to 0° Lat, and include the model/WMO results for the Tropical mean (as shown in Table 5)? Also, for the most poleward latitudinal bin, it looks like the “modeled” values are ploted at a higher latitude than the “observations”. This must be due to the total number of data-points used to compute the VSLS value. This should be explained either in the caption or in the text.

Figure 15: Why not including panel for WISE_TACTS in this figure?

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