Review ACP-2019-1091

Seasonal impact of biogenic VSL bromine on the evolution of mid-latitude lowermost stratospheric ozone during the 21st century

1 general comments

`evaluating the overall quality of the discussion paper`

In this manuscript, the authors study the impact of brominated very short–lived substances VSL^{Br} on the evolution of extratropical and tropical ozone in the lower stratosphere (LS).

The authors conducted two sensitive studies with and without an additional 5 ppt bromine source from VSLS in the stratosphere. An ensemble of three simulations for each scenario has been run from 1960s until the end of the 21st century with 10 years of spin-up from 1950–1960. Stratospheric halogen background loadings (e.g. CFCs, CH_3Br) follow the RCP 6.0 scenario. The model used was the CESM1. The model setup, forcing data, and experimental setup is described comprehensively.

This study complements and completes previous studies which focused on specific aspects or shorter/selective timespans and is therefore valuable in understanding the impact of VSL^{Br} by many means. The authors especially:

- deduct the efficiency of ozone depletion efficiency under additional bromine load from VSL with respect to pure CO_x, BrO_x, and mixed CO_x/BrO_x depletion and compare it to the efficiencies of O_x, HO_x, and NO_x induced ozone losses;
- comprehensively study the impact of seasonal variation in the VLS source strength on ozone depletion in the UTLS, and
- attribute these to seasonal variation in chlorine heterogeneous reactions.

All studies in this manuscript are conducted for mid-latitudes in both hemispheres as well as the Tropics. Hence, the current title limiting the results to "mid-latitude" is misleading, because the authors study not exclusively mid-latitudes. I suggest to adjust the manuscript title to account for this.

The scientific content of the manuscript is sound, there are only a few minor remarks. The language is overall concise but needs some refinement where the statements are not entirely clear.

2 specific comments

`individual scientific questions/issues`

Abstract

- L17/L18/L22: *"extratropical"/"extra-polar"/"mid-latitudes"* Without further information, these terms may be interchangeable. If the authors, indeed, refer to the same region (latitude band), they should decide on ONE of the terms and use it consequently throughout the text. In case different regions are meant, the terms need further explanation since exact definitions vary from article to article. (Only in Section 2 a definition for "mid-latitudes" is given.)

Section 1

- P3L20: *"high-mid latitudes"* Same as above – how do the authors define this region?

- P3L37-38: *"[...] i.e. with an interactive ocean [...]"* How does this study benefit from using an interactive ocean if the authors consider fixed seasonal VSL emissions to reduce uncertainty (see Section 2)? This information might not belong to Section 1 and should at least be mentioned or discussed at the appropriate place in Section 2.

Section 2

- P4L32-P5L3: While the configuration of the model is very well described in this section, the authors do not discuss the consequences (if any) the low-top and low stratospheric resolution has on their analysis. They should elaborate on this.

- P5L3-4: *"The current CAM-Chem version includes a non-orographic gravity wave scheme [...], an internal computation of the quasi-biennial oscillation (QBO) [...]"* Which CAM-chem version do the authors refer to here? The current release is CAM6.0. Previously, they wrote about CESM1 with CAM4 and cited Tilmes et al. (2016). The referred article states regarding the QBO: "The limited vertical resolution of the FR model configurations does not allow for the generation of an internal QBO in CAM4-chem. Therefore, for the FR CCMI experiments, REFC1 and REFC2, the QBO is imposed in the model by relaxing equatorial zonal winds between 90 to 3 hPa to the observed interannual variability, following the approach by Matthes et al. (2010)." The authors should clarify the version which they are referring to and in case they meant CAM4, rephrase the sentence in a way that it is clear that the model does not generate an internal QBO.

- P5L6-7: *"[...] this model configuration uses a fully coupled Earth System Model approach, i.e. the ocean and sea ice are explicitly computed."* Doesn't this introduce additional uncertainties due to, e.g., radiative and temperature feedback? On the other hand, these ought be strongest in the Arctics which are not subject to this study. The authors should include a brief discussion on the advantages/disadvantages of this ESM approach on their study.

- P5L8-10: *"Two independent experiments (each of them with three individual ensemble members only differing in their 1950 initial condition) [...]"* For the sake of reproducible, how do these initial conditions differ? On climatological time-scales do initial conditions even matter?

Section 3

- P6L22-27: *"As the oceanic* VSL^{Br} *emission [...]"* and the following *"Although the increase [...]"* The statement in the second sentence appears to be incomplete. With respect to the first statement (sentence), do the authors intent to say, that there are projected changes in atmospheric dynamics/chemistry (the ones they list) using the RCP 6.0 emission scenario suggesting a change in product gas injection (PGI), but they don't find these in their simulation? Or do they intent to say that the listed changes negate each other which is why they don't find any trend in PGI in their simulations? The authors should elaborate on this.

- P6L28: *"The validation of inorganic chlorine [...] clearly shows a good model-satellite agreement [...]"* The term "model-satellite agreement" may be considered as "lab-slang". One should rather write "good agreement between model and satellite" or "the model is in good agreement with satellite observations". Beside of this, it is not clear that the authors are referring to Fig. 1 as "validation".

- P6L32-34: *"The slight underestimation [...]"* If this is truly due to unaccounted anthropogenic chlorine sources, this means that the RCP 6.0 projections regarding future emissions of chlorinated species are erroneous. What are the implications on future ozone and climate? Based on their findings, the authors may discuss this in Section 4.

- P8L7-9: *"[...] introduces a continues reduction in TOC that exceeds the model ensembles variability between experiments [...]"* I might have missed it, but where is the "model ensembles variability" actually quantified in the manuscript?

- P8L18: *"[...] the largest model bias compared to SBUV [...]"* How does the model bias compare to the uncertainty in the SBUV data? If the authors cannot quantify this it should be at least discussed here.

- P8L20-21: *"[...] much larger ozone loss efficiency [...] within the mid-latitudes [...]"* Maybe point out that this is true in both hemispheres.

- P8L38-P9L2: *"[...] which shows a continuous decline in the course of the 21st century from its peak values observed during 2000 [...]"* As mentioned above, one has to be aware at this point, that decline this is due to the emission scenario for CFCs used in the model. As mentioned above and as the authors mentioned citing Hossini (2015b, 2017), if CFCs are not out-phased as demanded by the Montreal protocol or if new anthropogenic sources of halogenated substances occur in the future the projections will change. This should be, at least, mentioned in Section 4.

- P11L5-P12L22: This subsection's content is rather difficult to follow at times due to the abbreviations in use. Especially the terms, $\text{Halog}_{-\text{Loss}}$, $\text{BrO}_{\text{x}-\text{Loss}}$, and VSL^{Br} are confusing. There seems to be no clear seperation between in the term $\text{Halog}_{-\text{Loss}}$ and $\text{BrO}_{\text{x}-\text{Loss}}$ with and without additional Br from short–lived substances. I will come back to this later on. The authors may elaborate on their terminology in this regard.

Section 4

In addition to the point mentioned above, since the results for the mid-latitude are substantially different from the ones for the Tropics, the authors may consider to separate these into different subsections to make it more clear.

3 technical corrections

`purely technical corrections`

3.1 General

- Although it is a matter of personal style/taste: There are many preceding conjunctions throughout the text which appear to be fillers. The authors may consider to remove those.
- The authors use ^{o} throughout the manuscript instead of °. This should be corrected for.

3.2 Specific

Abstract

- L20: *"[...] the inclusion of* VSL^{Br} *result [...]"* result → results
- L33/L35 and others: *"The largest modelled [...] the spring [...]"* As long as not a specific spring or winter, etc. is meant, the prefix article is wrong.
- L36/L37-38: *"*$\text{Halog}_{-\text{Loss}}$*"* Just a remark: In the final typesetting this ought not be broken at the page's margin.
- L35-37: *"[...] the halogen-mediated ozone depletion [...] is more efficient than [...] respect to other seasons."* respect → with respect

Section 1

- P1L15-17: *"Owing to their short lifetimes, the impact of VSL on stratospheric ozone peaks at the extratropical lowermost stratosphere [...] an important atmospheric region [...]"* I suggest to insert "which is" → "[...] which is an important atmospheric region [...]" to make the sentence more readable.

- P1L34: imply → implies
- P3L2: *"[...] 3,290 ppt* Cl *and 19.6 ppt* Br *[...]"* Only a remark: It is rather uncommon to use ppt for such high concentrations (chlorine), but the authors probably intent to show the difference between chlorine and bromine concentrations in the most comprehensible way.
- P3L7: *"[...], understanding the role of natural* VSL^{Br} *sources is key for chemistry-climate projections."* Missing article in front of "key".
- P3L11/P4L22/P11L25: *"[...] has been reviewed elsewhere [...]"/"[...] has been given elsewhere [...]"/"[...] has been described elsewhere [...]"* Not sure if this is a proper citation style. Use "by" instead and have in-text citations?
- P4L5-9: *"The layout of the paper is as follows: Section 2 resumes the main characteristics [...]"* I assume "resume" is not the right word here. The authors probably meant "summarize".

Section 2

- P4L16-17: *"[...] with the exception that here we consider a constant and geographically distributed and seasonally-dependent oceanic emissions [...]"* Substitute "here" with "in this work" or something similar; emissions → emission.
- P4L18-19: *"It is worth noting that this emission inventory [...], which allows introducing a complete seasonal cycle on the emission strength [...]"* "It is worth noting that" can be dropped. As mentioned above, it merely serves as filler in this context; "allows introducing [...] on" → "allows to introduce [...] to".
- P4L22-23: *"Monthly and seasonally varying zonally averaged distributions lower boundary conditions of long-lived chlorine [...] were considered [...]"* I don't understand this sentence. Did the authors mean to write: "Monthly and seasonally varying zonally averaged distributions of long–lived chlorine [...] and bromine [...] were considered as lower boundary conditions [...]"?
- P4L22-28: *"Monthly and seasonally varying [...]"* This sentence is way to long. I suggest to break it up into two sentences. The natural breaking point would be L26 "[...], while [...]".
- P4L28-29: *"In order to avoid unnecessary uncertainties associated with the speculative evolution of VSLBr [...]"* The terms "unnecessary" and "speculative" are too judgmental in this context. You may rephrase to "[...] reduce uncertainties associated with the uncertain evolution [...]".
- P4L29-31: *"[...] we used a constant annual source strength for the whole modelled period [...]"* A "constant annual source strength" somehow implies constant emissions which is probably not what the authors intent to say based on the previous

sentences and the half-sentence in parenthesis which follows. It is clear that they treat the Ordoñez et al. emissions inventory as climatology, not projecting any future increase/decrease in emissions or changes in the seasonal cycle which they call "speculative". The authors should make this more clear.

- P4L32: *"The CAM-Chem configuration used here [...]"* here → in this work
- P5L21-22: *"[...], the zonal mean of the ensemble mean of each independent experiment (*run^{LL} *and* $\text{run}^{\text{LL+VSL}}$*) [...]"* The usage of the term "run" seems to be ambiguous throughout the text. On P5L10/L13, a run refers to one single model integration or set-up but in this sentence and later on it seems to refer to the ensemble mean? The authors should elaborate on this.
- P5L33-37: Maybe the use of a list could increase the readability of this paragraph?

Section 3

- P6L5: *"In addition, Figure 1 also [...]"* Either "in addition" or "also" is unnecessary in this sentence, because they mean the same in this context. Please drop one or the other.
- P6L7: *"[...] the temporal evolution [...] show [...]"* show → shows
- P6L14/L46: present-time → present–day; present times → present–day
- P6L19-22: *"Although within the tropical lower stratosphere [...], the contribution [...]"* Consider to split this sentence at "the contribution".
- P6L34: *"Moreover, although [...]"* Filler, conjunctions can be removed.
- P7L18-20: *"A clear example is the fingerprints [...]"* This sentence is not clear. Do the authors mean to say: That the fingerprints of the Indian summer monsoon can be seen in the $\text{ClO}_\text{x}/\text{Cl}_\text{y}$ ratio at the given location (15 °N and 100 hPa)?; is → are
- P8L5-6: *"A list of interesting features can be observed [...]"* This phrase sounds colloquial. Maybe change to "Many interesting features [...]". Talking about lists, the author may consider to actually use a list for their points (i)-(iv). This would increase the readability of the text and content at this point.
- P8L33: *"[. . .] the seasonal relatives [...]"* It is not clear, what the authors intent to say here, especially since "relatives" (= family members) is the wrong word. Do they mean the "relative difference in ΔTOC for each season"? This should be clarified.
- P8L34: *"dotted"* This is probably not the right term in this context. There are actually no "dotted" lines in Fig. 5. All lines are solid, but some (in the lower part of the plot) display gaps which indicate the significance of the changes. The authors may consider changing their wording here.

- P9L16-21: *"In contrast to mid-latitudes [...] for the tropics [...]"* The results for the Tropics are the total opposite compared to mid–latitudes. To not confuse the results, it would increase the readability if this paragraph was to begin in a new line.

- P9L32-33: *"In contrast [...] is at least half-fold [...]"* "half-folded" is most likely not the proper term in this context. Did the authors mean that ozone loss due to VSL^{Br} is reduced by 50 % by the end of the 21st century compared to present–day?

- P9L34: *"In agreement with our results, [...]"* This should be rephrased using "our results are in agreement with".

- P9L37: *"Interestingly, deepest O3(z) reductions [...]"* Drop "interestingly" if you don't mean to say that this is completely unexpected; here and similar at other places (e.g. P10L16): deepest → largest / greatest.

- P10L24-27: Some missing white spaces in front of or behind "-"; "Montreal Protoco" missing "l"

- P10L28: "This is line [...]" → "This is in line [...]"

- P11L13: *"[...] over the SG-ML lowermost stratosphere [...]"* over → in

- P11L30: seasonality increase → seasonal increase

- P11L37: *"crossed* $ClO_x - BrO_{x-Loss}$*"* Do you mean "mixed"?

Section 4

- P13L18: *"[...] winter. with maximum [...] reaching"* with → The; reaching → reaches

- P13L19-21: *"We find that the inclusion of* VSL^{Br} *leads to seasonal changes on the overall depth and vertical distribution of ozone [...], which could result to [...]"* This sentence is not clear. By "overall depth", do you mean the TOC?
changes on the → changes in the; results to → results in

- P13L30-33: *"[...] entirely dominated by* BrO_{x-Loss} *[...]"* Also in Section 3 and elsewhere in the manuscript, the authors use the term BrO_{x-Loss} to indistinguished between ozone depleted by BrO_x from long–lived opposed BrO_x from short–lived substances (VSL^{Br})? This is somewhat confusing, as already mentioned above, since all organic bromine has to be transformed into inorganic bromine to deplete ozone.

Figures

The authors should, in general elaborate on:

- be more concise in captions. It is not always easy to see (without reading the whole caption) what each panel actually refers to since the relevant description is embedded in long sentences.
- choose a different color map for most of their contour plots since the "rainbow colors" imply visually a distinct divergence of data at the edge between blue and green/yellow. This may lead to unintended misinterpretation[1]. It is therefore depreciated, but unfortunately still widely used.

Fig. 1

- With respect to the most common type of color-blindness, the authors may refrain from using green together with red in plots when there is no difference in line style or luminescence of the colors.

  Caption

- L3-4: *"(a) global upper stratosphere (6.5 hPa) and lower stratosphere (50 hPa) (b) tropics [...]"* The way it is written, it looks as if "lower stratosphere [...]" is part of panel (a) instead of (b)-(d). The authors should make this more clear. Maybe use ";" instead of "and".
- L5: *"show changes"* Wouldn't it be use the term "evolution" for consistency?
- L7: *"The red triangles of all panels [...]"* You can drop "of all panels".
- L9: dotted → dashed

Fig. 2

See comment about color map.

Fig. 3

Panel labels (a)-(i) move around quite a bit. Would it be possible to set them to lower left corner for each plot?

Fig. 6

- The y-axes' ranges changes between the plots ((a,c) 300–4; (b) 100–4; (d,e,f) 300–5) hPa. Please make the range consistent in all of the plots.
- Panel (f): See above comment on color map.

  Caption

- L2: within of → within the
- L3: shows → show

Fig. 8

- The axes' lines are far thicker than the actual markers or lines symbolizing the data which is distracting. Partly due to this the violet line and especially the black line at the very bottom in panel (a) are easily missed. Please reduce the axes thickness and think about to change the color for O_{x-Loss}.
- The legends are overlapping with the content and should be placed outside of the plot area (right hand side).
- The plots (b) and (c) are extremely busy and not well explained in the caption. Why are data for JJA displayed using dashed lines in contrast to the thin, solid line/marker combination used for the remaining "seasons"? You should rather use either four different line styles or markers. Please make this consistent.
- Is it possible to show the actual ensemble standard deviation in this plot? It is mentioned in the text but nowhere quantified.

  Caption

- L4-6: *"The panels (b) and (c) show the seasonal mean contributions [...]"* This is not concise. Already is it difficult to spot LL and LL+VSL in the plots' legends, but the way this is presented in the caption is not making it any easier to interpret this plot. The actual information in what (b) and (c) differ comes in the very end of a long sentence. A better approach would be, e.g. "Seasonal mean contributions of [...] families for the different experiments (b) run^{LL+VLS} and (c) run^{LL}."

Fig. 9

- See comment about color map above.
- The color bar indicates an open interval in both direction (blue and red triangles). If the data actually is confined in a closed interval between 0 and 100, the triangles should not to be used.
- Can you show or quantify the magnitude of difference between the percental contribution of BrO_{x-Loss}, $BrO_x - Cl_{x-Loss}$, and ClO_{x-Loss} in both, run^{LL} and run^{LL+VSL}? One cannot draw this information of a plot with color levels of widths 10 % especially for panels (c,d). (Even worse in Fig. S9).

Supplementary figures

All comments above apply to similar figures displayed in the supplement.

References

[1] D. Borland and R. M. Taylor Ii, *LaTeX: Rainbow Color Map (Still) Considered Harmful*, IEEE Computer Graphics and Applications, vol. 27, no. 2, pp. 14-17, March-April 2007.