

Review of the manuscript “Atmospheric impacts of chlorinated very short-lived substances over the recent past. Part 1: the role of transport” by Bednarz et al., ACPD, 2022.

The paper presents a modeling study using a whole atmosphere CCM to evaluate how sensitive is the stratospheric loading of chlorinated very short-lived species (Cl-VSLS), including both Source Gas Injection (SGI) and Product Gas Injection (PGI), to different model configurations for the dynamical transport (i.e., considering free-running and nudged setups). The analysis focused on the Cl-VSLS evolution during the 2000-2019 period, where they found an overall SGI enhancement from 40 ppt to 80 ppt for the free-running simulation, and up to 10-20 ppt additional Cl-VSL when the model was nudged to ERA-Interim or ERA-5 reanalysis. The larger SGI for the nudged configurations resulted in an overall smaller SGI + PGI due to the faster transport. To evaluate the total inorganic chlorine evolution in the stratosphere, they present a comparison of HCl and COCl₂ trends in the stratosphere and upper troposphere with satellite observations (ACE-FTS). They show that regardless of the transport configuration, the inclusion of Cl-VSLS improves the model performance, although the hemispheric asymmetry observed in the lower stratosphere is only captured with the nudged simulations. The work is very well planned and provides a realistic and clear evaluation of the magnitude of the Cl-VSLS contribution to the total inorganic chlorine loading in the lower stratosphere. The methodology and results are generally well presented, although some clarification is required as described below. I suggest the paper is accepted for publication after the following issues have been solved:

Main Comments:

1. Splitting the project in Part 1, Part 2, Part 3.

The authors decided to split the paper in 3 parts, but what each of the parts is about is only clear in the last sentence of the conclusions (P12,L371-375). I suggest making it clear since the beginning to avoid the reader wondering him/her-self about what the other parts will address. Note that in P6,L186, the authors explicitly mention that the HCl comparison with satellite data will be presented in a following paper, but it is included here in this draft (Section 4).

2. Year-to-Year variability for individual free-running ensemble members

I found it very surprising the very small variability on the SGI (as well as for the SGI + PGI) among the 3 individual members of the free-running ensemble (Fig. 3). I wonder how large is the year-to-year variability for the free-running simulations, in comparison with the difference between the free-running mean and each of the nudged simulations? For example, during the

first 5 years after the spin-up (when the Cl-VSLS LBCs remain almost constant), the year-to-year variability for a single ensemble is much larger than the variability between the free-running individual simulations during a single year. Then, could this year to year variability in the free-running simulations be more representative of the model variability than computing the multi-ensemble mean?

In addition, when reading section 4.3 (Fig. 9, rightmost panels) it is surprising that HCl trends for the different ensemble members present such a large variability, while the Cl-VSLS variability between ensembles from Fig. 3 seems to be very small. Have you computed the Cl-VSLS trends using the same procedure (and in the same units as for the HCl trends, % per decade) to evaluate if the SGs are also showing a large range of trends for each ensemble-member as for the PGs?

3. The role of Chemistry representation

The impact of transport on the total SGI is explained in detail. However, the sum of SGI + PGI is only briefly discussed in Section 3 (P7,219-224). In particular, the individual PGI contribution is not addressed for any of the configurations, and no individual number is given. It is only mentioned that the trends and variability of the SGI + PGI are smaller for the nudged simulations than for the free-running. Even though I understand the authors focus the product gas discussion on Section 4 and 5 where they compare with HCl and COCl_2 observations, I believe the paper would benefit of extending a bit the discussion of the overall PGI before moving on the Age of Air trends. Unless the PGI is going to be presented in detail in Part 2 of the paper. In addition to i) the enhanced transport for the nudged simulations (P6,L222) and ii) the faster large scale circulation in the stratosphere (P6,L223), the change in PGs abundance in the upper TTL can affect the washout efficiency of halogenated species and therefore modify the overall PGI (see Fernandez et al., 2021 for the case of bromine).

Minor Comments:

P5,L160-162: What is the mean value for SGI for the final year 2019? Because by looking at Fig. 3 it looks smaller than 80 ppt for the annual mean. The text mentions a couple of times that the Cl-VSLS is “doubled” ... just wanted to be sure if is not “almost doubled”.

P7,L227-P8,L233: It is not clear how the seasonal mean for MAM and SON (and the other seasons, none of them are defined) was computed. Furthermore, the text mention that the

model output was deseasonalised and then explain that the seasonal mean was used. I suggest re-order and re-phrase to make it clear.

P8,L249: “a strong asymmetry in its horizontal structure”. Do you mean a hemispheric asymmetry?

P8,L259-P9,265: Section 4.2 begins mentioning that “nudged simulations … show similar interannual dynamical variability to observations”, and later concludes that “such pattern is thus similar to that found in the ACE-FTS data”. The analysis is correct, but first, it should be “more” similar (not just similar), and second it is not clear if the initial sentence points at the AoA comparison until it is later explained in the paragraph. I suggest re-ordering.

P10,L317: Any explanation on why COCl_2 shows a much smaller variability than HCl for the same set of ensembles?

P11,L345-346: It should be made clear that the sentence applies for the nudged simulations.

P12, L366: What do you mean by “which effectively constitute only one ensemble member”?

Language editing comments and Typos:

P2,L58: Engel and Rigby et al., 2018 (here and elsewhere).

P3,L88; P4,L118-120, P8,L235: Consecutive points and/or double-spaces.

P5,L159-160: “The stratospheric source gas injection (‘SGI’) of chlorine from Cl-VSLs can be approximated based on their simulated concentrations at 17 km and 25°S-25°N”. The term can be approximated here can introduce confusion as someone can infer that you did not computed it this way. I suggest to rephrase.

P6,L166-170: The sentence is confusing: at the beginning you said that “experiments have, by definition, different meteorology”. But later it is mentioned that “ensemble members are forced with identical chemical (Cl-VSLs) and meteorological (SST and sea-ice) LBCs”. I suggest using oceanic/surface LBCs at the end of the sentence.

P6,L185: What do you mean by “brought about from the inclusion”?

P6, L191: Move “more CI” before the opening parenthesis to make it clear what the 20 ppt are.

P8,L229: Define ACE-FTS

P8,L251: Fig. 7g → Fig. 8g.

P9,L264: Fig. 7c-f → Fig. 8c-f.

P9,L270-274: Make sure you point at the proper panels of Fig. 9 for the tropics, NH and SH regions.

P10,L300;P10,L311: Fig. S6 → Fig. S7.

P11,L334: Is it “sense” or “sign” ?

P18,L568: Remove Sturges et al., 2000 at the end of the reference.

Figures and Tables

Figure 1: The figure shows the CI-VSLs surface LBCs. I understand this is an “input” to force the model at the surface and not an “output” of the model. If that is the case then instead of “simulates” it could be changed to “forced” or something similar this.

Figure 2: “Annual mean zonal mean” reads awkward. Also note that CI/VSLs in panel e and CI_tot in panel f are not defined.

Figure 3 (also Fig. 7 and elsewhere). Note that the degree symbol is printed erroneously on the pdf file.

Figure 3 caption: At the very end of the caption it should be “SGI + PGI”. Make it explicit in the caption that the 3 individual members of the free-running ensemble are shown.

Figure 10: I found it surprising that AoA trends for panels a and b showed such a different distribution. Does it impacts on the analysis? Should it be highlighted in the text?

Supplement:

Affiliations are missing. They should be made consistent with the main text.

Fig. S1: missing → mixing

Fig. S4: It should be made clear in the caption that results are for each ensemble

Fig. S6: main paper → main text.

References

Engel, A., M. Rigby, J.B. Burkholder, R.P. Fernandez, L. Froidevaux, B.D. Hall, R. Hossaini, T. Saito, M.K. Vollmer, and B. Yao, Update on Ozone-Depleting Substances (ODSs) and Other Gases of Interest to the Montreal Protocol, Chapter 1 in Scientific Assessment of Ozone Depletion: 2018, Global Ozone Research and Monitoring Project—Report No. 58, World Meteorological Organization, Geneva, Switzerland, 2018.

Fernandez, R.P., J.A. Barrera, A.I. López-Noreña, D.E. Kinnison, J. Nicely, R.J. Salawitch, P.A. Wales, B.M. Toselli, S. Tilmes, J.-F. Lamarque, C.A. Cuevas, and A. Saiz-Lopez, Intercomparison between surrogate, explicit and full treatments of VSL bromine chemistry within the CAM-Chem chemistry-climate model, *Geophys. Res. Lett.*, 48(4), doi:10.1029/2020GL091125, 2021.