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Review of 'Atmospheric impacts of chlorinated very short-lived substances over the recent past. Part 1: the role of transport' by Bednarz et al.

Anonymous Referee #2

Referee comment on "Atmospheric impacts of chlorinated very short-lived substances over the recent past – Part 1: Stratospheric chlorine budget and the role of transport" by Ewa M. Bednarz et al., Atmos. Chem. Phys. Discuss.,
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Bednarz et al. present a study exploring the transport of Cl-VSLS into the stratosphere in both free running and nudged version of their chemistry-climate model. I found this study to be very interesting, exploring an important topic in the contribution of Cl-VSLS to recent stratospheric chlorine changes. The comparison of the free running and nudged simulations nicely demonstrates the sensitivity of Cl-VSLS injection to model dynamical fields. The paper is clearly written and laid out and the figures are of high quality. However, while I found the analysis presented in the paper to be fine, the framing of that analysis and the conclusions reached in the paper should be tempered, particularly the conclusions regarding the sensitivity to choice between free running and nudged model configurations. I also feel that as the study is part of three planned manuscripts, more should be said earlier in this manuscript about what will be covered here and what elsewhere. The title of this manuscript implies that wider atmospheric impacts will be evaluated, and it is only clear in the final paragraph that changes to ozone will be considered in a follow-up. I would recommend publication of the manuscript after the authors address the comments below.

General comments:

The paper is titled atmospheric impacts of chlorinated VSLS, but there is very little evaluation of the impacts of Cl-VSLS on wider atmospheric chemistry/chemistry-climate coupling in the paper. Instead, the paper is really an evaluation of the impact of Cl-VSLS on stratospheric chlorine, particularly HCl. This is probably because the authors plan to publish their work in three parts. But it is only made clear at the end of the manuscript what will be included in the other parts. The authors should move this information to much earlier in the manuscript. I also feel sentences like 'Atmospheric impacts of chlorinated very short-lived substances over the recent past (up to and including the year 2019) were assessed using the UM-UKCA chemistry-climate model', as used in L321-322 should be qualified by making it clear the authors in this study are only looking at the impact of VSLS on stratospheric halogens, and the sensitivity of this to model dynamics.

An argument that is made by the paper that I'm not sure I agree with is that model results are very sensitive to the choice to run the model in free running or nudged modes, i.e., the model configuration. But presumably this arises because there are biases/differences between the modelled dynamics compared to the reanalysis datasets. If so, I feel the argument should be changed to 'our results highlight the importance of model dynamical fields on transport of Cl-VSLS into the lower stratosphere'. In a model that has dynamical fields that look very similar to the reanalysis datasets, the authors could have concluded that there is very little sensitivity to the choice to run the model in free running or nudged modes. Otherwise, if the authors are arguing that the process of nudging the model is introducing additional tendencies into the transport of Cl-VSLS species that simply do not exist in the free running version of the model, or that this different configurations fundamentally change composition-dynamical coupling/feedbacks in such as way as to significantly influence the modelling of Cl-VSLS, then I don't feel the authors show enough to conclude this in this study.

The DEST chemical scheme used in the paper is cited as in prep. Is this still the case? It seems to me that this scheme is key to producing and interpreting the results presented in the paper, but the reader doesn't know what it is, or how it models stratospheric halogens. If the paper has not yet been published, I would encourage the authors to include much more information on what is in the DEST scheme here.

Is it possible to add the tropopause to all the latitude height cross sections? The focus on the paper is the transport of Cl-VSLS into the stratosphere, and it would aid the reader to show where that is in this model.

Specific comments:

L22-31: I found it difficult to follow the arguments in this paragraph. The abstract states that in 2019 there is 80 ppt of stratospheric Cl from source gas injection, and an additional 50 ppt from product gas injection, for a total of 130 ppt. The authors then state that nudging the model to ERA-I meteorology results in a 20 ppt increase in lower stratospheric Cl-VSLS source gases, which is equivalent to a doubling. I feel this paragraph needs to make it clearer where in the stratosphere these numbers are applicable, and to better relate the values in the free running simulation to those in the nudged simulation. It is clearer in the main text, but on first reading I was comparing a change of 20 ppt to the 130 ppt earlier in the paragraph, and so confused by the doubling comment.

L39: The effectiveness of the Montreal Protocol is better measured by changes to the gases it controls, e.g. CFC-11 mixing ratios. Trends in HCl are of course affected by this, but also by changes to atmospheric transport (as discussed in this paper) and stratospheric chlorine partitioning as CH4 and N2O etc change.

L48: controls seems too strong here – CO2 and water vapour both play important roles in dictating stratospheric temperatures. Similarly 'controlling' on L50, halogens are of course important, but HOx, NOx and Ox cycles a also key to accurately modelling stratospheric ozone.

L49-50: Does ozone not absorb longwave radiation at all altitudes?

L169: I do not feel that meteorological LBC is a good description of SSTs and sea ice boundary conditions and would suggest this is changed.

L189-195: I feel care has to be taken when discussing these values. Given the strong gradients in this region, the way the values are currently presented is misleading, and need further clarification. At the level of the 20 ppt contour in figure 5c, the values have approximately doubled as the authors state, but at the level of the 40 ppt contour, which is within a few km of the 20 ppt contour, the increase is ~50%. Presumably above the 20 ppt contour the percentage increase in Cl-VSLS could be very large.

L197: Given the increase seems rather uniform over a range of latitudes, I wonder to what extent might tropopause height changes contribute to the mixing ratio changes seen here? Increased tropopause height may also contribute to the younger age of air seen in Figure 6.

L233: Presumably this is because the ERA-Interim data ends on 31st August? Why not calculate all the trends over the same time period? It seems odd to include the 2019 SON mean in all but one of the panels.

L299: The authors use the tropical average at earlier points in the paper, but only define it as 30S-30N here. I would move this definition to earlier in the manuscript.

L321-325: I feel this paragraph is summing up all three parts of the planned study, two of which are not yet available. Can this study claim to be an end-to-end assessment if there

is no discussion of stratospheric ozone changes, impacts on stratospheric temperatures, coupling between the composition and dynamics, and wider atmospheric feedbacks?

L351-352: Care should be taken here – presumably if the modelled dynamics were more consistent with those in the reanalysis datasets the authors would not make this claim. See my general comment above.

Technical comments:

L51: Replace 'so far' with 'to date'

L61: I would add 'comparatively' before 'few studies' – there is now a large and growing body of literature looking at chlorinated VSLS.

L119: 2 fullstops

L203: ERAI-Interim -> ERA-Interim. Also L214

L204: lager -> larger

L206: al -> al.