

## AUTHORS RESPONSE TO REVIEWER #1

The paper presents a modeling study using UM-UKCA with and without CL-VSLS halogens oriented to evaluate the impact of chlorinated very short-lived substances (VSLS) on recent-past stratospheric ozone trends (2010-2019). In addition, a new estimation of the ozone depletion potential (ODP) and stratospheric-ozone depletion potential (SODP) of the dominant CL-VSLS species (dichloromethane) is provided. The simulations are properly designed, the paper is well written and organized, and the analysis of the results is coherent. Their main conclusion is that CL-VSLS induce a modest but non-negligible role on the stratospheric ozone budget that counteract some of the gains achieved by the Montreal Protocol, but that the inclusion of CL-VSLS in UM-UKCS does not considerably modify the magnitude of diagnosed ozone trends in the tropical lower stratosphere. I suggest the paper is accepted for publication after the following issues have been solved:

We thank the reviewer for positive review and helpful comments that have improved the manuscript. We address the individual points below in blue.

### Main Comment:

**Update the comparison with other modeling studies** The authors properly compare their main results with previous publications performed by the group and/or co-authors, which are in line with the results presented here (i.e., recent ozone trends in the lower stratosphere are dominated by dynamics and not chemistry). However, a discussion with other results showing a significant contribution from VSLS chemistry (e.g., Villamayor et al., 2023) would improve the manuscript (see specific comments below). In addition, the discussion would benefit of a comparison of CL-VSLS impacts on ozone trends with respect to that arising from BR-VSLS (e.g., Sinnhuber and Meul, 2015; Barrera et al., 2020).

We thank the reviewer for pointing this out – we have now modified the manuscript to include more discussion of the role of Cl-VSLS for ozone trends (Section 4), including comparison of trends inferred from nudged vs free-running simulations, as well as comparison with the recent study of Villamayor et al. (2023). We have also added some comparison with the impacts from Br-VSLS (as reported in the study of Barrera et al., 2020) into the discussion (Section 6).

### Minor Comments:

P1,L17-19: Are the values provided in the abstract “annual mean” or “springtime mean”? In case different time-averaging apply, please make it clear.

We have now made this clear.

P2,L54-57: You should relate this with the recent publication by Villamayor et al. 2023 that found that inclusion of both natural and anthropogenic VSLs in a CCM results in a significant chemical signal that contribute to ozone reduction in the low-latitudes lower stratosphere.

We have now changed this part to read:

“While the effect of Cl-VLSL on the tropical lower stratospheric ozone trend in a chemistry-transport model has been estimated to be small (Chipperfield et al., 2018), a larger impact has recently been reported using a global chemistry-climate model containing a coupled troposphere-stratosphere chemistry scheme including chlorine, bromine and iodine VLSL (Villmayor et al., 2023), and as such the issue should still be re-examined.”

P3,L74-75: It would be interesting to include some time of comparison between FR and SD. For example, a panel like Fig. 1c but presenting the mean +/- sigma of the latitudinal variation of ozone changes for the FR ensemble simulations. This would help to evaluate how well the FR range compares with the SD-runs values?. Note that Fig. 3 in part-1 of this paper (Bednarz et al., 2022) shows that FR simulations have a larger SGI, but similar SGI+PGI than SD simulations ... Thus I would expect a larger O3 influence for FR simulations. Is that the case?

Unfortunately, some of our output got corrupted, and as such we do not have the data to produce the corresponding changes in total column ozone in the free running VLSL and BASE simulations. We have, however, calculated the corresponding yearly mean changes in ozone concentrations analogous to Fig. 2a, and included it as Fig. S2 in the Supplementary Material. We have also added the following paragraph to Section 2:

“Given the significant dynamical variability characterising ozone levels on year-to-year timescales, we focus in this section on the results from the nudged model simulations. Whilst the corresponding free-running UM-UKCA simulations suggest higher Cl-VLSL induced lower stratospheric ozone losses (Fig. S2), consistent with the larger Cl-VLSL product gas to source gas stratospheric chlorine injection (Bednarz et al., 2022), there is large uncertainty in these values due to the contribution of natural variability.”

P3,L80-81: What do you mean by “warm winters”? Are those 2-3 DU differences observed in Winter? or in Arctic spring preceded by warm winters? In addition, note the sentence only apply to high-latitudes, and is confusing as it mixes "warm winters" with "on average each year". Please rephrase and expand to make it clear.

We have now rephrased and clarified this part; we also now don't use the term 'warm winters' at all.

P3,L81-81: Smaller over the Arctic, but larger over SH high-latitudes. Why is that? Due to the larger VSL-CL emissions in NH? Or could temperature changes play also a role here?.

We apologize for the confusion. We have discovered a numerical problem with one of the simulations used (BASE<sub>SD5</sub>) and have now re-run the simulation and updated the plots in the manuscript. We note that the correction does not substantially affect the conclusions of this paper, or the results of its accompanying PART 1 (Bednarz et al., 2022, <https://doi.org/10.5194/acp-22-10657-2022>). There is now, however, a much better agreement in the average Cl-VLSL induced ozone between the two sets of nudged simulations (Fig. 1c that the reviewer is referring to).

P3,L86-87: Similar to P3,L74-75, you highlight the importance of nudging to evaluate the impact on Arctic results, but I wonder how large the difference between SD and FR simulations can be.

See the response to P3, L74-75 above.

P4,L108-109: What is the rationale for showing ClO values at the beginning of march but O3 values at the end of march for nudged simulations? To evaluate the "cumulative" role of VSL-Cl chlorine on O3 losses?

We indeed chose to show ClO values at the beginning of March but O3 values at the end of March to evaluate the cumulative role of Cl-VSLs on ozone loss. We also note that Arctic ClO values tend to peak in late winter/early spring, while ozone losses tend to be largest later on in the season.

Such an approach has been previously used in many published studies of Arctic ozone depletion (e.g. Manney et al., 2011, doi:10.1038/nature10556; 2020, doi:10.1029/2020GL089063).

P5,L135-136: As described, the chemical signal is negligible when SD are considered ... but induces an additional 0.5-1.0% enhancement for FR simulations (Fig S3). The authors may want to evaluate this and comment in more detail.

We thank the reviewer for the useful suggestion. We have now expanded Section 4 to include more discussion of this; we have also added the middle panel of the old Fig. S3 that shows the tropical ozone trends in each simulation into the main manuscript (now Fig. 5).

P5,L138-139: The results are in line with Chipperfield et al., 2018 and in contrast to Villamayor et al. 2023 ... which include both natural and anthropogenic VSLs. Note that Villamayor found also a negative trend, although not significant, when only VSLs-Cl were considered.

As per response above, we now include more discussion (including references to the recent Villamayor et al. study) in the manuscript.

P5,L143-144: I suggest writing the numbers in the text as mean +/- sigma or mean +/- 2sigma ... as the range is already shown in Table 1.

Corrected.

P5,L151-152: I suggest mentioning explicitly that Cl-VSLs are not considered in the Montreal Protocol and/or amendments.

Thank you – we have now included this.

P5,L155-156: This absolute numbers for chlorine could be compared with VSL-Br impacts in the lower stratosphere as described by Sinnhuber and Meul, 2015 and Barrera et al., 2020.

We have now added the suggested comparison with the Br-VSLs impacts:

“In comparison, the ozone loss from the natural brominated VSLs emissions during the same time was estimated at ~1-2 DU in the tropics and ~5-6 DU in the midlatitudes (Barrera et al., 2020), albeit using a different climate model.”

P6,L156-157: Results summarized here apply mostly for the nudged simulations (SD), but not for the FR. A comment on this would be helpful.

We now clarified this in the text.

P7,L192: I'm confused about the LBCs description: I thought for the case of CH<sub>2</sub>Cl<sub>2</sub> and C<sub>2</sub>Cl<sub>4</sub> an emission inventory (Claxton et al., 2020) was applied instead of using LBCs. Could you please make this clear?

Section M1 of the Methods is correct, i.e. LBCs of all four Cl-VSLs are used in these transient simulations as a source of Cl-VSLs (instead of emission inventory); this is the same approach as used by the PART1 of this study (Bednarz et al., 2022, <https://doi.org/10.5194/acp-22-10657-2022>). While we have performed simulations with CH<sub>2</sub>Cl<sub>2</sub> and Cl<sub>2</sub>Cl<sub>4</sub> emissions following the emission inventory of Claxton et al. (2020), these simulations are not used in either this manuscript or its companion PART1, but instead will form a future follow up PART3 study. We apologize for the confusion.

P7,L203-204: Here you mentioned a source, but above you mentioned only LBCs were used. Please make it clear. In addition, if you provide a "perturbation value" here, it would be nice to provide the "base CH<sub>2</sub>Cl<sub>2</sub> emission value" in previous paragraph, so one can easily estimate the magnitude of the perturbation. Similar to CFC-11, could you please provide the surface LBC for the base simulation.

We apologize for the confusion – we now clarify all of this.

#### **Language editing comments and Typos:**

P1,L16: Remove period "." after "time."

Corrected.

P4,L105: "shows that up to 25 ppt of the elevated ClO" or "shows differences up to 25 ppt for ClO". Please revise and make it clear.

Corrected.

P5,L158: What do you mean by "exact results"?

We have clarified this as the results in particular regions and seasons.

P7,L198: replace "ed" by "performed" or similar.

Corrected.

P8,L217: is it "where" or "when"

Corrected.

P8,L219: replace "included" by "including"

Corrected.

## Figures and Tables

Figure 1: The final sentence of the caption should say annual global mean "total ozone column" changes ... similar for the "total ozone column change" magnitude that is missing in the Y axis of panel c.

Corrected.

Figure 2: Please use "ozone difference (%)" instead of the way it is written. Also I suggest including the dynamical tropopause line or 150 ppbv chemical tropopause line to help the reader splitting the troposphere and stratosphere.

Corrected, and the tropopause added.

Figure 3: Why do you show Ozone differences for 31 March but ClO differences for 1 March?

As discussed above, Arctic ClO values tend to peak in late winter/early spring, while the ozone losses tend to be largest later on in the season (when the cumulative impact of halogen chemistry is largest). We note that such an approach has been previously used in many published studies of Arctic ozone depletion (e.g. Manney et al., 2011, doi:10.1038/nature10556; 2020, doi:10.1029/2020GL089063).