

AUTHORS RESPONSE TO REVIEWER #2

Scientific Significance: The scientific questions addressed in this paper are appropriate for ACP. This work addresses for the first time the recent (2010-2019) impact of chlorinated Very Short-Lived Substances (VSLS) on ozone. The authors state they found “modest and non-negligible role of Cl-VSLS” to the stratospheric ozone budget. They also emphasized that continued Cl-VSLS emissions “could offset some gains by the Montreal Protocol”. They were the second group to “estimated” the ODP of dichloromethane. I highly recommend this work for publication. Below are a few comments that may add to the impact of this work.

Scientific Quality: This work is of high scientific quality. The authors use a state-of-the-art CCM (UM-UKCA) nudged to ERA-Interim and ERA-5 meteorology. They also show results from an ensemble mean from the UM-UKCA CCM. The Cl-VSLS chemistry is also represented in a detailed manner.

Presentation Quality: Generally, in good shape. However, I would increase the font size of Figures 3 and 4.

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

Specific Comment.

Abstract, line 16. Typo “for the first time. Using the”

Corrected

Abstract, lines 19. I found it confusing in the abstract when the authors highlight the 2011, 2014, and 2020 years and state that up to 5-6DU monthly and zonal mean Arctic ozone reductions are simulated. Then in line 20 they highlight year 2020 with “~6DU ozone in total by the end of March”. They state 2020 was a recent cold winter. I would suggest reworking sentences here being more specific why you picked 2011 and 2014 relative to 2020?

We apologize for the confusion. We have discovered a numerical problem with one of the simulations used (BASE_{SD5}) 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), but it does reduce the Cl-VSLS induced ozone loss inferred from the runs nudged to ERA5 in 2014. We have now changed the text accordingly.

Abstract, lines 21-23. The authors state that Cl-VSLS “do not considerably modify the magnitude of the recent ozone trends”. Please be more specific, is this is tropical, polar, global, everywhere, etc? Also, why would one expect the trend to be significant over a short period (i.e., 2010-2019), especially in the Arctic? I would suggest adding more detail in the abstract if you want to mention ozone trend results.

We apologize for the confusion. While that particular statement in the abstract is fairly generic, i.e. relate to ozone trends in general in most of the stratosphere, the focus of our

study is to explain the persistent, statistically significant negative ozone trends diagnosed from the observations in the extra-polar lower stratosphere (Ball et al., 2018; 2019).

We have now clarified this in the abstract: “Despite ~doubling of Cl-VSLS contribution to stratospheric chlorine over the early 21st century, the inclusion of Cl-VSLS in the nudged simulations does not substantially modify the magnitude of the simulated recent ozone trends and, thus, do not help to explain the persistent negative ozone trends that have been observed in the extra-polar lower stratosphere.”

Abstract, line 22. The ODP of Cl-VSLS is quantified. The paper mentioned this was the second ODP derivation. What do you mean that it was “estimated” – is that a typical way to discuss the derivation of an ODP? This topic deserves a couple sentences to clarify why you feel it is important to put this discussion in the abstract. See my comment on lines 146-147 below.

We have now change ‘estimated’ to ‘calculated’, as well as added the following discussion to Section 5: “ODP is an important and well-established metric that is reported in WMO/UNEP Ozone Assessment Reports and other policy-facing documents to gauge the possible ozone depleting effect of a gas relative to CFC-11. Unlike for long-lived species, there are few explicit (i.e. based on global model calculations) ODP estimates of VSLS in the literature. This in part reflects the relative complexity of a VSLS ODP calculation, which requires consideration of both the source gas and product gas injection of halogens to the stratosphere. A sensitivity of the ODP to emission location and season can also play a role for some species (e.g. Brioude et al., 2010). Given the significant upward trend in the CH₂Cl₂ production and emission from its predominantly industrial source, the quantification of ODP for CH₂Cl₂ is particularly important.”

Lines 46-49. “We showed that the contribution from these Cl-VSLS to stratospheric chlorine had increased from 70 ppt Cl in 2000 to 130 ppt Cl in 2019, i.e. almost doubling over the first two decades of the 21st century.” One could make an argument that this information was taken from Bednarz et al., 2022, Part 1 – but it would be nice have this information brought to the abstract level when summarizing the trend results.

We agree and have now included this in the abstract.

Lines 53-54. In addition to Chipperfield et al. 2018, Wargan et al., 2018, and Orbe et al., 2020, Stone et al. also came to this conclusion (that dynamical variability is driving the O₃ trend) using a chemistry climate model similar to UM-UKCA. Stone, K. A., Solomon, S., & Kinnison, D. E. (2018). On the identification of ozone recovery. *Geophysical Research Letters*, 45. <https://doi.org/10.1029/2018GL077955>.

We have added the reference to the Stone et al. study.

Line 85-86. “Furthermore, no significant Cl- VSLS-induced Arctic ozone loss can be diagnosed from the model ERA-Interim nudged monthly and zonal mean data for the spring 2011; this might be related to the small size of the polar vortex in that year and thus difficulties in reproducing its dynamical properties in a nudged model setup.” This sentence is a bit disconcerting in that the reader is meant to figure why there are difference in the choice of reanalysis products. The main question in my mind is why even show ERA-Interim in this study? Presumably ERA-5 is the best ECMWF product to look at nudged ozone trends?

We agree with the reviewer. However, in our previous work (PART1, Bednarz et al., 2022) we explicitly discussed the role of the choice of reanalysis used for nudging for the simulated Cl-VSLS stratospheric input and the stratospheric chlorine budget. As such, we believe it is important to include the results made with both reanalysis datasets for completeness.

We have now expanded the discussion as to the possible reasons behind the differences in the simulated ozone responses: “We note that while very similar average large scale ozone losses are diagnosed from the simulations nudged to different reanalysis products (Fig. 1c), some differences can emerge for individual regions and seasons. In particular, no significant Cl-VSLS-induced Arctic ozone loss is diagnosed for the spring 2011 from the model nudged to ERA-Interim, while the Arctic ozone loss modelled in the spring of 2014 is notably higher in that run than in the run nudged to ERA5. This might be related to the generally small and variable size and structure of the NH polar vortex, thus difficulties in reproducing its dynamical properties in a nudged model set up, or to the differences in the resolved transport between the two reanalyses (e.g. Diallo et al., 2021; Ploeger et al., 2021; Bednarz et al., 2022). These results thus suggest that the choice of reanalysis for nudging could also be important in some years for the diagnosed ozone impacts from Cl-VSLS.”

Figure 3 and 4. Please increase the font size of the titles and x-axis.

Done.

Lines 114-118. This is a very interesting discussion, i.e., “the impact of curbing emissions of long-lived ODSs achieved by the Montreal Protocol was estimated to reduce the magnitude of the Arctic ozone depletion in that spring by up to ~35 DU in mid-March compared to peak halogen levels in early 2000 (Feng et al., 2021).” It might be useful to bring this comparison of curbing the emissions of long-lived ODSs achieved by the Montreal Protocol up to the abstract level (i.e., versus 6 DU from Cl-VSLS)?

We agree that this is an interesting and relevant discussion. We note, however, that the result of Feng et al. was derived using a different climate model (i.e. TOMCAT/SLIMCAT chemistry-transport model), and as such a close abstract-level direct comparison may be misleading without sufficient amount of details. We now clarify the use of a different climate model in the text.

Lines 146-147. “The calculated stratospheric ODP of 0.0102 (confidence interval of 0.0062-0.0163) is similar to the whole atmosphere ODP metric, implying that CH₂Cl₂ has a relatively small effect on ozone below the tropopause in UM-UKCA.” This is an interesting result. Is there anything more you can say about this result? Is this due to where CH₂Cl₂ is emitted (e.g., China)?

We have now added the following discussion of this result to Section 5: “In part, this reflects the relatively long tropospheric lifetime of CH₂Cl₂ (~100 days in the boundary layer; Hossaini et al., 2019), especially compared to some particularly short-lived iodine species (e.g. CF₃I) for which the distinction between ODP and SODP can be particularly important (Zhang et al. 2020).”