Comment on acp-2021-880
Anonymous Referee #2

*We thank the referee for the thoughtful comments and suggestions; we address these (in italics) in the text below under **"Answer"**, for each point made. Our revised document will clearly include the changes we point to in detail below.*

Referee comment on "Upper stratospheric ClO and HOCl trends (2005–2020): Aura Microwave Limb Sounder and model results" by Lucien Froidevaux et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-880-RC2, 2021.
Review of the manuscript ACP-2021-880 by Froidevaux et al. submitted to ACP and entitled "Upper stratospheric ClO and HOCl trends (2005-2020): Aura Microwave Limb Sounder and model results.

This manuscript presents satellite measurements and model simulations over 50S-50N for the time period 2005-2020 for two inorganic chlorine reservoirs, namely chlorine monoxide (ClO) and hypochlorous acid (HOCl). These global measurements characterize the upper stratosphere with a vertical resolution of 3-4 km (ClO) and 5-6 km (HOCl), they have been derived using the Optimal Estimation Method from Aura-MLS radiometric observations that have been consistently gathered over 16 years (i.e., without the hardware failures that have affected other MLS products/channels, e.g., HCl, N2O). Online and offline products are used, and they present a very good sampling, with about 3500 profiles available per product and per day.

This places the authors in a good position for robust trend determinations for difficult targets, especially HOCl, in support of the Montreal Protocol on substances that deplete ozone. To this end, they use a method or approach that accounts for the auto-correlation often present in geophysical data series, and their model further includes proxies for parameters known to affect the abundance of stratospheric tracers (the solar cycle, the QBO, …).

The investigations are supported by model simulations performed by the WACCM model constrained by MERRA-2 meteorological fields. These simulations are available at the observation sampling, and they are analyzed in the same way than the observations to provide ClO and HOCl climatologies and trends. The model is also used to perform a sensitivity study in order to determine the dependence of the simulated HOCl distributions to the assumed kinetics for its formation reaction ($ClO + HO_2 \rightarrow HOCl + O_2$), these parameters being still insufficiently defined, and possibly responsible for significant satellite-model biases.

All these investigations are conducted with great care and the results are well presented. The manuscript is well organized, and all the figures are clear, self-explanatory and useful. The text is sometimes a bit lengthy, for instance in section 4 (discussion), but this is probably more a matter of taste than a real issue. It is important to note that this study is dealing with inorganic chlorine reservoirs for which only few observations are available (and the measurements are challenging!), its publication would therefore bring original and interesting elements to the community, also in the context of the continuous evaluation of the success of the Montreal Protocol. In my opinion, this manuscript is almost ready for publication, and I only have a few suggestions to bring to the attention of the authors.

Primary suggestions or questions.

In the introduction (line 73), the authors remind us that mid-term variability complicates the trend detection in the lower stratosphere. Among others, Strahan et al (2020) have reported about hemispheric asymmetry in stratospheric transport trends and its impact on the distribution of long-lived tracers. This question is absent elsewhere in this manuscript and global trends are consistently reported. Moreover, most of the material/figures presented do not allow the reader to make his/her own opinion about possible upper stratospheric signals that could characterize the observed and modeled ClO and HOCl data sets. The time series are restricted to a Northern/Southern belt for ClO/HOCl (Fig. 2/8). Still, Fig. 3 and 4 exhibit some asymmetric signals, e.g., at 31.6 hPa for the first one, or the "light green bubble" seen for ClO in the Southern hemisphere in the 20-40 S and 2-10 hPa ranges in Figure 4. My bet is that the authors decided to report global trends because it is already challenging enough, especially for HOCl. But still, I would suggest to indicate in the introduction and/or conclusion that there were no signs of asymmetry in these upper stratospheric data sets, or that they were not searched for.

Answer: *We agree that we could add a few more sentences regarding the issues of asymmetry, given what we already can actually see in the trend Figures that are provided (and these trends follow the time series variations). Doing more than this would require further work, and maybe more importantly, such studies are probably better pursued with time series from longer-lived species (and into the lower altitude portion of the stratosphere, which could not be done in this work on MLS ClO and HOCl). Indeed, underlying lower stratospheric variations in these short-lived species will be significantly influenced by variations in longer-lived species such as CH_4 and H_2O. At present, we cannot say too much more, but in section 3.1, end of the 2^{nd} paragraph, we have added the following sentences: "We note (from Figs. 3 and 4) that there is some asymmetry in the stratospheric ClO trends between the two hemispheres, with stronger decreases at northern than at southern midlatitudes, and with a somewhat more pronounced effect in the lower stratosphere. However, these asymmetries do not carry much statistical significance. These tendencies are opposite to what has been observed in HCl column trends (see Strahan et al., 2020), which show more rapid (longer-term) declines in the South than in the North. Lower stratospheric ClO trends are likely to also be related to trends in CH_4 and H_2O, although we do not pursue this quantitatively here, other than through the WACCM results, which show a similar, but slightly stronger interhemispheric lower stratospheric ClO trend asymmetry than in the MLS data. At 32 hPa, we note that there is evidence for low frequency (multi-year) MLS and model ClO variations with poorer regression fits to both data and model (although not shown here, and not the focus of this work); this complexity is a likely reason for the larger trend discrepancies (WACCM versus data) in this region. Further investigations of lower stratospheric interhemispheric trend asymmetries (and related age of air issues) are probably best pursued through detailed studies of longer-lived species than ClO."*

Also, in the Conclusion section, we have summarized this by adding the following two sentences at the end of the 2^{nd} paragraph: "Between 15 and 32 hPa, there are indications of some interhemispheric asymmetry in the MLS ClO trends, with faster decreases at northern than at southern midlatitudes, although this is not statistically significant; there is also evidence for low frequency (multi-year) variability, especially at 32 hPa. Further investigations of lower stratospheric interhemispheric trend asymmetries (and related age of air issues) are probably best pursued through detailed studies of longer-lived species than ClO." We did not feel that we could *just add two sentences in the Conclusion section without having discussed this, even briefly, in the text. We thank the referee for this suggestion.*

In section 2.2 (line 190), it is indicated that the WACCM6 runs have been augmented to cover the more recent years than the initially available simulations. But since the boundary conditions for the halogenated source gases have been provided by a reference published a few years ago (Meinshausen et al., 2017), I wonder which data have been used in order to describe the post-2016 evolutions of the source gases? A more recent reference is probably needed here.
Answer: *For completeness, we have added a reference (Meinshausen et al., 2020) to the list at the place mentioned by the referee, but more importantly, we note that the CMIP6 scenario is used to <u>project</u> GHG and organic halogen <u>inputs</u> for the model <u>beyond 2014</u>, so this added (underlined) wording should clarify the approach, to a large extent regardless of exact references, or reference dates.*

In section 4 (starting line 505), the authors indicate that changes in upper stratospheric temperatures should have had little effects on chlorine partitioning, with less than a 1K decrease as observed by Steiner et al. (2020) over the period of interest here. Regarding the partitioning and trends derived from WACCM6, I guess that the (small) temperature change is accounted for thanks to the MERRA-2 meteorological fields? Or in other words, is the temperature trend in MERRA-2 in agreement with the ~1K trend of Steiner et al. (2020)?
Answer: *Yes, the WACCM6 temperatures follow the MERRA-2 inputs, and MERRA-2 assimilates observational inputs discussed by Steiner et al. (2020).*

Figure 3: I would suggest here to swap the axes (latitude for x-axis; trend for y-axis). This way, Figure 3 would report the latitude on the horizontal scale, as is the case for Fig. 1, 4 and 6.
Answer: *Thank you, we have followed this suggestion; we have also changed the similar Figure (S3) for HOCl in the Supplement.*

Minor point

In the introduction (line 59), I would also mention the effect of photolysis (in addition to transport and mixing) to explain the conversion from tropospheric chlorine into the reservoirs.
Answer: *Agreed, we have added the word "photolysis" to the explanation.*