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Comment on acp-2022-151

Anonymous Referee #1

Referee comment on "The influence of energetic particle precipitation on Antarctic stratospheric chlorine and ozone over the 20th century" by Ville Maliniemi et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-151-RC1, 2022

This paper investigates the influence of energetic particle precipitation on ozone and chlorine in the SH stratosphere over the 20th century by means of SOCOL3-MPIOM chemistry climate model simulations. EPP-induced NOx increases and associated ozone decreases were found to be in agreement with results of previous studies. A new finding is that EPP also induces substantial ClO decreases in the upper and mid stratosphere which reduces the ozone-depleting efficiency of EPP. In the lower stratosphere, EPP-induced ClO increases and ozone decreases were obtained at the end of the century while the opposite occurred before the period of high chlorine load. These results suggest a significant modulation of EPP-induced ozone loss by atmospheric chlorine which has implications for the future evolution of polar stratospheric ozone. This is a relevant topic and the paper is certainly suitable for publication in ACP.

The paper is well written, however, it fails short in convincingly identifying the chemical processes that are responsible for the EPP-induced ClO changes. Regarding the upper atmospheric response, the authors note in the abstract that the ClO decreases go along with increases in chlorine nitrate. A closer look at the absolute changes in the chlorine partitioning, however, suggests that most of the ClO is converted into HCl rather than into ClONO2.

Regarding the lower stratospheric response, the only explanation for the encountered ClO response is that "ClO is increased by activation of chlorine from the reservoirs" in the presence of PSCs. This is well known but does not explain why the ClO increase is enhanced by EPP. A possible reason for enhanced chlorine activation under EPP could be that the ClONO2-limited heterogeneous processing on PSCs in the Antarctic lower stratosphere is accelerated by the availability of more NOx and hence ClONO2.

It is also unclear why the lower stratospheric ClO response changes sign around the 80ths with increasing chlorine load. Is it possible that associated ozone depletion alters the Cly partitioning which could then modulate the ClO and O3 responses? Low ozone favors HCl formation and reduces ClONO2 by increasing the NO/NO2 ratio through the NO+O3 reaction (which then increases the rate of the ClO+NO reaction).

In summary, a more detailed analysis of the chorine partitioning in absolute terms (i.e. by use of line plots of the seasonal evolution of ClO, HCl, and ClONO2 from both EXP and REF simulations at 10 and 100 hPa levels for both low and high chlorine load conditions) would be very useful for identifying the responsible processes which, in turn, would significantly enhance the strength of this paper.

Therefore, I strongly encourage the authors to address these points in a revised version, together with the specific comments listed below.

l162-163: I agree that the ClONO2 decrease under EPP in mid-winter, seen in Fig. 8, suggests a Cly partitioning in favor of HCl by reaction R9. However, there is essentially no NO in the dark polar mid-winter stratosphere which could react with ClO. Although there could be a minor NO contribution from the the sunlit region, it is still striking that the ClONO2 decrease occurs in mid-winter and not in spring when sunlight (and hence NO) is available in the entire region.

l168-169: Webster et al. (1993) looked at an Arctic winter which might not be representative for the Southern hemisphere. In any case, an explanation about *how* EPP reduces the HCl amount is missing.

l180 "ClO-ClO catalytic cycle". Maybe "ClO dimer cycle" is more common.

l181ff / Fig. 9d: The EXP-RED TCO difference is negative throughout the winter/spring. This is in contradiction to the observational results of Gordon et al. (2021) who showed a TCO increase during SH spring (Oct-NOv) in high EPP years.

l190-193: The change of sign in the ClO response around the 80s is particularly interesting in Fig 10b. However, this is not discussed in the manuscript.

l193-198: It is unclear how the discussion on GCR/EEP/SPE helps to understand the negative ozone response in the last two decades of the century. All these types of EPP produce NOx. The key questions are: Why is the lower stratospheric ClO response positive at the end of the century (it is negative in the middle and upper stratosphere...)? Why does it change sign with the onset of enhanced chlorine load?

l198: "ClO seen in the lower stratosphere". Do you mean "ClO response seen in the lower stratosphere"?

l213: "is more than expected". Do you mean "is more than unexpected"?

l221: "We propose that this ClO increase can be explained by activation of chlorine from reservoir species ClONO2 and HCl." This is well known. What needs to be explained here is the positive ClO *response to EPP* after 1980.

l231: What do you mean with "ideal simulations"? Idealized model experiment? If yes, what kind of experiments?