

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2022-151

Anonymous Referee #2

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-RC2>, 2022

This paper investigates the impact of energetic particle precipitation (EPP) forcing of chlorine species, and its consecutive impact on EPP-NO_x driven stratospheric ozone loss. Ensemble model runs with and without EPP are carried out over the whole 20th century (1900-2008), a period with high solar activity and high chlorine loading in the second half of the 20th century. The impact of particle precipitation on NO_y, HO_x and ozone in the middle atmosphere has been studied in detail in a number of publications, but analyses of the impact of EPP on chlorine species are rare; the publication thus provides a new aspect. Of particular note is their observation that the high chlorine loading apparently had an impact on stratospheric ozone loss due to EPP, presumably by restricting both NO_x- and ClO_x-driven catalytic cycles due to the reaction of ClO with NO₂. The inference is that in the coming decades, when the atmospheric chlorine loading will decrease, EPP ozone loss via NO_x catalytic cycles will likely become more efficient. The paper is generally very well written, and the conclusions appear sound. However, conclusions could become more robust with a few more analyses, see suggestions below. Also it seems to me that the EPP ClO_x mostly transfers into HCl, not ClONO₂, and a more detailed discussion of this, and of possible pathways, would be useful.

Lines 54-56, R8 and R9: include and discuss pathways of HCl formation in the introduction, as this appears to be important as well: HOCl + Cl → HCl + ClO; HO₂ + ClO → HCl + O₃; OH + ClO → HCl + O₂, anything else? This works via an increase in HO_x; EPP HO_x is available during the particle precipitation in the (upper) mesosphere, but also possibly due to storage of EPP NO_x and EPP HO_x in the form of HNO₃, which is transported down into the stratosphere during winter and there slowly photolyses, releasing both NO_x and HO_x (Verronen and Lehmann, GRL, 2015)

Line 89: wouldn't it be more exact to use only data from REF for the estimation of the significance? Than (EXP-REF) would be tested against the variability of REF, which seems to be more to the point.

Line 104: maybe you could say a few more words about the content and meaning of Fig 2.

Figure 3 – for better readability, please include ticks for 10 hPa and 0.1 hPa for the vertical axis. Same for Fig 4 and following.

Line 117: if the mesospheric ozone depletion is due solely to in-situ EEP HOx production, why is it stronger in the Southern hemisphere? Doesn't the difference between Northern and Southern hemisphere imply a dynamical/long-lived component in the mesospheric ozone depletion as well? Possibly HNO₃ formation/photolysis?

Line 120-121 and following discussion of lower stratosphere ozone anomaly: the positive ozone anomaly covers nearly the whole lower stratosphere, from high Southern to high Northern latitudes, with the exception of the polar winters, when the anomaly turns sign. In mid-and low-latitudes, this positive anomaly is interpreted by the authors as a GCR impact (line 123), and this appears likely. However, I would argue based on the spatial/temporal evolution of this signal that this GCR signal extends from pole to pole, but is overwritten by the auroral signal indirect effect during polar winter.

Line 129: Are these corresponding to the negative NO_x anomalies? And, is there a corresponding anomaly of ClONO₂?

Line 135: Over hundred percent --> more than a hundred percent

Line 143: ... as can be seen in Fig 2a --> by comparing with the Ap index shown in Fig 2a. However, it would be better to provide some hard numbers here to substantiate this statement, e.g., by providing a correlation coefficient (preferably from some ordered, non-linear method – rank? – not Pearson) between NO_x and Ap.

Fig 7 b and e, as well as following figures – can you provide error bars due to ensemble variability for the timeseries?

Fig 7 c and f: the lines appear to be anti-correlated – are they? E.g., provide correlation coefficient

Line 147: Loss of ... as this is a decrease relative to the reference scenario, I wouldn't call it "loss", which would imply chemical loss

Line 151: seems to be anticorrelated --> just provide the correlation coefficient

Line 154: response --> response to EPP forcing

Lines 154-156: and possibly because NO_x is bound in PSCs in the form of HNO₃?

Line 158-159: Substantial increase compared to what – EXT to REF, or to the beginning of the model periods?

Line 161: the ClONO₂ amount is not negative in your model runs (one hopes), it is less than in the REF scenario without EPP.

Line 163: also strengthened by the fact that the ClONO₂ difference in absolute numbers seems to be much smaller than the HCl difference. I think you could explore this in more detail. Do you really think this is due mainly to CH₄ + Cl?

Line 173: again, just provide a correlation coefficient

Figure 8 c and f: are the lines anticorrelated? Are they correlated/anticorrelated to the Ap index shown in Fig 2a?

Line 215: We find a significant decrease of stratospheric ClO ... relative to a model run without EPP impact / due to the EPP impact

Line 219: ClO abundances decrease ... relative to a model run without EPP .. by ...

Line 220: Why is this negative before 1980?

Line 230: In principle, I agree with this conclusion, but find "crucially" maybe a bit too strong / confident.