Reply to interactive comment on "Semi-empirical models for chlorine activation and ozone depletion in the Antarctic stratosphere: proof of concept" by P. E. Huck et al.

Anonymous Referee #1

General comments
The manuscript introduces two semi-empirical models to describe the evolution of the vortex averaged ClOx and ozone mass deficit (OMD). The authors show that their simplified but physically based models in combination with statistical fitting are able to capture the observed variability of ClOx and OMD during the last 30 years. The subject of the manuscript is relevant to the ACP scope and potentially interesting for the community, because it could provide an inexpensive way to forecast the behavior of the considered quantities in the future. I think the publication of the manuscript can be recommended. However, there are several issues in the manuscript (see below) and maybe some moderate revisions will be necessary before the publication.
We would like to take this opportunity to thank the reviewer for taking the time to provide this feedback on our paper.

Specific comments
1. The authors use FAP which is based on the temperature threshold for NAT PSC formation, but the liquid aerosol seems to be more important (e.g., Wegner et al., 2012, ACP) and the authors should briefly discuss this issue. The ICE PSC also should play an important role in the formation of ozone hole; however its contribution is missing in the proposed parameterization. Does the good agreement with observations means that these processes are not important for the ClOx and OMD evolution?
FAP is meant as proxy for the geographical extent of conditions favourable for chlorine activation. It is derived from meteorological analysis based on a threshold temperature for the existence of such conditions. The particular numerical values used in this study result from using 195K as the temperature threshold. The formation of STS is a gradual process without such a clear threshold. But the efficiency of chlorine activation on STS is a steep function of temperature and 195K is very close to the small range of temperatures at which the chlorine activation on STS becomes efficient on timescales of days or hours. So using 195K as proxy for chemical processing due to heterogeneous reactions is meaningful even though the real process of activation is much more complicated and involves different types of droplets or particles in the stratosphere. Because the FAP time series essentially acts as a basis function in a regression model, as long as the shape of the time series is appropriate, including the start and stop times, the amplitude of the time series is less important since this would be compensated for by a change in the magnitude of the fit coefficient.

2. The authors state: "Heterogeneous reactions on PSC surfaces lead to the conversion of reservoir forms of chlorine into active forms (Solomon, 1999, and references therein). Once reactive chlorine is exposed to sunlight, chlorine-catalysed ozone destruction begins. . .". However, the heterogeneous reactions do not lead to immediate chlorine activation. During the first stage Cl2 reservoir is formed and the presence of sunlight is necessary to convert it to ClOx. Potentially, the Cl2 formation is possible for FAS=0 and ClOx formation could act even if FAP=0. Does it have any implication for the proposed parameterization? The equation (1) will not produce any ClOx if these two processes are sequential, and even for the time periods when FAP=0. I think it would be helpful to show the applied evolution of FAP and FAS from eq.1 at least for selected time periods.
While it is true that in this method the heterogeneous chemistry processes have been simplified such that ClOx formation cannot take place if FAP=0 or FAS=0, FAS is never zero since there is always some region of the vortex exposed to sunlight. FAS therefore simply becomes an intra-seasonal weighting factor for FAP, giving greater weight to FAP-driven increases in ClO_x later in the season compared to earlier in the season. Therefore the product of FAS and FAP is only zero when FAP is zero. But note that it is not this product that drives ClOx – it drives dClOx/dt. As a result, when FAP is zero, ClOx is not zero but decays according to the rightmost term in equation (1). Therefore this construction of equation (1) does not have any implication for the proposed parameterization. We have now included a figure showing FAP and FAS from 1992 to 1997.

3. The amount of Cly in the proposed parameterization does not depend on the actual meteorological situation, however it does in reality due to interannual variability of vertical descend and cross vortex mixing. In turn, these quantities depend on the ozone sate (especially for the future climate). What implications for the forecast could have

such a simplification?
The reviewer is correct that this simplification can lead to inter-annual variability not being tracked completely. The simplifying assumptions generate an additional source of uncertainty. This may account for why in some years the model does not track the observations perfectly (see Figure 3). A term could be added to equation (1) so that Cl_y was e.g. some function of vertical descent and cross-vortex mixing. This may well improve the quality of the fits. But it also creates an additional requirement on the semi-empirical model when it is being used in prediction mode i.e. we would now need projections of inter-annual variability in vertical descent and cross-vortex mixing. Where-ever these come from, they would be just one possible realisation of the future. Output from the same model run with different initial conditions would produce a different but equally plausible Cl_y time series. To keep the model simple, but still useful, we require that the projection of Cl_y used as input to the model includes any *forced* (as opposed to that caused by internal variability of the climate system) secular changes (e.g. if there was a sustained increase in the strength of the Brewer-Dobson circulation), but consider meteorologically-induced inter-annual variability to be simply part of the 'noise' that is not tracked by the model. Since this noise cannot be projected anyway, we do not see this as a shortcoming of the semi-empirical model approach.

4. The authors suggest that the removal of NOx is parameterized using FAP, but it should be noted that NAT particles are too small and substantial sedimentation takes place only if they are inside ICE particles, therefore the irreversible denitrification could be substantially overestimated.
Our semi-empirical model uses (1-FAP) as a proxy for gas phase NO_x that deactivates ClO_x. But it is not necessary for sedimentation to occur to remove the NO_x from the gas phase. We make the assumption that if the vortex is completely filled with PSCs (i.e. 1-FAP=0) then there is no NO_x available to react with ClO_x to reduce ClO_x. For small quantities of PSCs the removal *rate* of ClOx increases because NO_x is now available. At no stage in this process do we make any assumption about whether or not sedimentation is occurring.

5. The authors suggested three processes responsible for the ClOx deactivation, but only one is included in the parameterization. What about other processes? Their intensity depends on the presence of the sunlight which is not included in the equation (1).
We have tested the equation with an additional term to account for ClOx deactivation depending on sunlight. The fitting result could not be improved significantly and therefore we decided for the simpler version of the model with fewer fitting parameters. Our goal is not to replicate every process in the system. If that were our goal we would eventually converge on a state-of-the-art chemistry transport model. Our goal is to emulate the bulk behaviour of ClOx and ozone with the fewest number of terms to produce a semi-empirical model with predictive capability that requires a small number of readily available input time series.

6. It is stated that:"The model tracks the observations for activation and deactivation of ClO well". It would be useful to provide some quantitative measure how well is the agreement.
We have added the information that 60% of the variance in the ClO observations can be explained by the model

Is it superior compare to the CCMs performance? The opposite trends in the observed and parameterized maximum ClO values are visible from the Fig.1. Is there any explanation of this behavior?
We have not compared these results with CCM output. The timing of activation and deactivations is represented by the semi-empirical model. The maximum is underestimated in the two earlier years, but considering the small number of data, we don't think a trend can be determined. A comparison against CCMs would be subject to the climate in the CCMs and there may be a significant trend in temperatures in the CCMs which would drive a trend in ClO that would not be indicative of what is happening in reality.

Minor comments and technical corrections:
1. Page 28456, line 2: I would use "over" instead of "from".
This has been corrected in the revised manuscript.

2. Page 28456, line 21: Which CCM were used? Probably the model should be briefly introduced.
The models UMETRAC and NIWA-SOCOL were used for this study and this information has now been included in the manuscript.

3. Page 28457, line 1: I do not understand why ". . .only temperature fields are required..".
Cly is also necessary.
Cly has been added to this sentence in the revised manuscript.

4. Page 28458, lines 21-26: The description of Fact is not satisfactory.
The equation for Fact has been added to the revised manuscript.

5. Page 28460, line 16: How H2O increase will promote formation of NAT particles?
For ICE particles it can be real, but they are not included.
It is true that increases in H2O in the stratosphere could affect the formation of PSCs (whether NAT or ICE or both). The key question for our semi-empirical model approach is whether a long-term secular increase in H2O would drive an appreciable increase in FAP since it is only through FAP that PSCs have any effect on the model. Our model has no sensitivity to whether the PSCs are NAT, ICE or both. Increases in water vapour could result in a small decrease in the 195K threshold for PSC formation assumed in our model. This in turn could affect FAP values. However, we expect this effect to be very small. More importantly however, it would be difficult to derive a reliable proxy for future changes in Antarctic lower-stratospheric water vapour that could be used as input to the semi-empirical model when it is being used in prognostic mode. For these reasons we have considered this to be a second-order effect which is excluded from our model.

6. Page 28460, line 16-18: This statement should be supported by a proper reference.
I do not think the sentence is completely correct.
The sentence has been modified in the revised version of the manuscript and the reference below has been included.
Reference:
Austin, J., and R. J. Wilson (2006), Ensemble simulations of the decline and recovery of stratospheric ozone, J. Geophys. Res., 111, D16314, doi:16310.11029/12005JD006907.

7. Page 28460, line 19-21: This sentence should be extended to give the readers an impression how it can be done.
For example, the equation can be fitted to two different temperature time series. The change in the fit coefficients could indicate which processes increase/decrease for different temperature scenarios. This information has been added in the revised manuscript.

8. Reference list: In my version there some extra numbers (they look like page numbers) at the end of references.
Thank you for pointing this out.

9. Figures: why not to show ClOx for 2000-2010?
We could show the ClOx time series but we have no observations to compare them to and so it would be just output from the semi-empirical model. We do not think that such a figure would add much value to the paper.