

Review of revised version of

“Future Arctic ozone recovery: the importance of chemistry and dynamics”

by E. M. Bednarz

General Comments

The authors have done a very thorough job revising their paper. They have taken great care of taking all the comments by all the reviewers into account. I have a few remaining comments, partly also on newly added material that I would suggest the authors take into account when revising their paper for publication in ACP.

The authors have added an analysis of the halogen induced ozone loss in the model vs VPSC (new Fig. 5). I think this is a valuable addition to the paper. The authors obtain different slopes of VPSC against chemical ozone loss for different time periods due to changes in the stratospheric halogen loading (as expected). There have been attempts to construct a variant of VPSC (PACL, Tilmes et al., 2007) than includes the changing halogen loading of the stratosphere in the definition. This quantity has been applied to an evaluation of heterogeneous processes in the polar lower stratosphere in the Whole Atmosphere Community Climate Model (WACCM3) for the period 1960—2003. It would be interesting to see the behaviour of the simulated ozone loss against PACL and see if the different slopes would collapse onto one curve. It would be good to add this aspect to the paper.

There was also a lot of discussion in the reviews about the representation of polar chemistry in general and heterogeneous processes in particular in the UM-UKCA model. The authors have explained now their equilibrium NAT scheme, where NAT is formed at the NAT equilibrium temperature, which I think is acceptable for this paper. However, it is known since a long time that NAT does *not form* at equilibrium so I would argue it should no longer be a “common technique employed in CCMs”. An aspect that I did not mention in the first review is the parametrization for reactivity on NAT that is used in the model. There are two competing and rather different formulations for NAT reactivity (Carslaw et al., 1997a,b); as NAT is important in the heterogeneous chemistry used here it might be worth clarifying this point in the paper.

Further, the heterogeneous reaction $\text{HCl} + \text{ClONO}_2$ is not included on liquid surfaces. But it is well known to occur on liquid surfaces (e.g., Hanson et al., 1994), so what is the reason for not including it? Possibly, the effects of a too early onset of NAT chemistry and the neglect of $\text{HCl} + \text{ClONO}_2$ on liquid surfaces cancel out to some extent. There is no point in changing anything here with regard to the manuscript in question. However, I suggest correcting these two issues in future model versions, i.e. include $\text{HCl} + \text{ClONO}_2$ on liquid surfaces and introduce a supersaturation requirement for NAT formation.

The details of the heterogeneous chemistry could however matter for predictions of the future; for example it is stated in the paper now that: “Stratospheric H_2O and HNO_3 levels are projected to increase in the future, which is likely to enhance levels of PSCs” However it depends on the type of PSC in question in how far an increase of H_2O or HNO_3 will affect chlorine activation. Nonetheless, the halogen induced polar ozone loss in the model will be in many respects robust against assumptions on heterogeneous chemistry (e.g. NAT vs. liquid; Solomon et al., 2015, see also discussion and references in my previous review). As this aspect is relevant for the model results presented here, I suggest discussing the issue briefly in the paper. For example on p 5, l. 5 you could briefly state not only that Keeble et al. (2014) obtained a reasonable representation of springtime Antarctic ozone, but also why. Perhaps something along these lines ~ “Previous studies (refs) obtain a consistent and relatively realistic representation of springtime Antarctic ozone for rather different assumptions on PSC formation so that the details of the heterogeneous chemistry in the model runs presented here should have no strong impact on the simulated halogen induced ozone loss. Moreover, we compare here the time evolution of polar ozone in a consistent framework, so that possible model biases should be less important”. Just a suggestion.

Finally, an important point in the reviews was also the discussion of chemical losses inside and outside the polar vortex. And a comparison between the 2060 and the 2063 model case. I see that the authors have responded in detail to this point and have modified and improved the manuscript accordingly. I think the analysis conducted for this discussion would also be very helpful to the reader (and not only to the reviewer/editor) and should be added to the manuscript. Perhaps in the supplement, but I suggest adding table 1 and Figs. R4 and R5 (or at least the left-hand columns of R4 and R5) to the actual paper together with the accompanying discussion from the reply (which then needs perhaps be a bit

extended/reformulated). But the material is there and I would recommend showing it.

In summary, I think the authors have done a very careful job in replying to the reviews and in revising the paper. I believe some further improvements are possible as outlined above. With these modifications, which should not be too difficult to do, I recommend the paper for publication in ACP.

References

Carslaw, K. S. and Peter, T.: Uncertainties in reactive uptake coefficients for solid stratospheric particles – 1. Surface chemistry, *Geophys. Res. Lett.*, 24, 1743–1746, 1997a.

Carslaw, K. S., Peter, T., and Müller, R.: Uncertainties in reactive uptake coefficients for solid stratospheric particles – 2. effect on ozone depletion, *Geophys. Res. Lett.*, 24, 1747–1750, 1997b.

Hanson, D. R., Ravishankara, A. R., and Solomon, S.: Heterogeneous reactions in sulfuric acid aerosols: A framework for model calculations, *J. Geophys. Res.*, 99, 3615–3629, 1994.

Solomon, S., Kinnison, D., Bandoro, J., and Garcia, R.: Simulation of polar ozone depletion: An update, *J. Geophys. Res.-Atmos.*, 120, 7958-7974, 10.1002/2015jd023365, 2015.

Tilmes, S., Kinnison, D., Müller, R., Sassi, F., Marsh, D., Boville, B., and Garcia, R.: Evaluation of heterogeneous processes in the polar lower stratosphere in the Whole Atmosphere Community Climate Model, *J. Geophys. Res.*, 112, D24301, doi:10.1029/2006JD008334, 2007.

Technical issues

Throughout the paper: change “statistically insignificant” to “not statistically significant”

On page 3., l. 7: you have changed “low temperatures” to “cold temperatures”; I would suggest the opposite. It is not that important (and “cold temperatures” is frequently used), but really only air can be cold not temperatures.

Correct this citation, the correct spelling is: Wegner, T., Grooß, J.-U., von Hobe, M., Stroh, F., Sumińska-Ebersoldt, O., Volk, C. M., Hösen, E., Mitev, V., Shur, G., and Müller, R.: Heterogeneous chlorine activation on stratospheric aerosols and clouds in the Arctic polar vortex, *Atmos. Chem. Phys.*, 12, 11095-11106, doi:10.5194/acp-12-11095-2012, 2012.