Review of "Uncertainties in modeling heterogeneous chemistry and Arctic ozone depletion in the winter 2009/2010" by Wohltmann et al.

This manuscript deals with the uncertainties in modelling heterogeneous chemistry and ozone loss in the Arctic winter 2009/2010. In order to test the uncertainties, the authors have performed a number of model runs by applying various chemical and physical assumptions. I find this study very interesting, as it gives an overview of possible uncertainties in the ozone loss calculation using chemistry transport modelling. However, I have some specific comments on some of the discussions presented in the manuscript. Please consider them for revising the manuscript.

MAIN POINTS:

1. Abstract needs to be revised with the uncertainty figures presented in the discussions. It should also include the main conclusion of various experiments performed. The impact of the uncertainties, as stated in the title, should be specified. For instance: please write the range of ozone loss values you obtained from the sensitivity runs and give the uncertainty range with respect to the reference run. Please also give a more appropriate concluding statement in the abstract.

2. You have used a lot of measurements from MLS and ACE-FTS. Please give a separate section for this. Description of data sets (accuracy, precision, vertical and horizontal coverage, biases) in the discussion sections detracts the attention of readers. Therefore, it would be good to give this section before you describe your model.

3. Please write a few sentences about the selection of the winter 2009/2010 for your case study here. May be you have a special reason for this (e.g. very cold first half of the winter).

4. Section 3.1 was described without relevant figures. This section should be modified with appropriate references. Note that the meteorological situation of this winter was well described in more than a couple of publications (please see this special issue publications).

5. You need to compare your results (ozone loss, denitrification, etc) with other published ones to generalise your findings. This should also be done for ClO comparison. There is at least one publication (for the winter 2009/2010) to compare with your results (see comments below).

SPECIFIC COMMENTS:

Page 26247, Lines 11—14: "Considerable changes in chemical evolution" do not affect ozone?

Page 26247, Lines 14—17: Please write the implication for this. Also, state whether you recommend any specific scheme to be used in CTMs in terms of your results.

Page 26248, Lines 1, 9: Write the relevant publications, if these are not personal communications.

Page 26248, Line 10: "current low estimate of ", please give a reference for this.

Page 26248, Line 22: Please specify that the possible bias in the "meteorological analyses used to force the model runs".

Page 26249, Line 1: SOLVE THESEO, VINTERSOL, EUPLEX need to be expanded. Please give reference for these campaigns. You could give the overview papers of these campaigns (e.g. Newman et al. 2001 for SOLVE).

Page 26249, Line 17: MLS does not measure PSCs. Please clearly state what you mean by this sentence (e.g. comparison of gas-phase HNO3).

Page 26249—26250: Model overview: Merge first three paragraphs.

Page 26250, Line 5: No numerical diffusion or comparatively less numerical diffusion ?

Page 26250, Lines 13—15: Delete this line. It gives a simple reason and a negative message. Instead, state that Sander et al. (2011) also give similar results.

Page 26253, Lines 11—14: Please reformulate this sentence.

Page 26253, Lines 21—22: Merge these two sentences to make a single paragraph. Replace Drdla and Müller with their ANGEO paper (see references). Please introduce the table properly.

Page 26254, Lines 25—28: Please give the region, altitude and time period of this bias.

Page 26255, Lines 17—19: Not to show, but to check the impact of unrealistic changes. Otherwise, you are predicting the results even before launching the model simulations.

Page 26257, Line 15: For wave 2 calculation, please give references here (e.g. Kuttippurath and Nikulin, 2012 and Dörnbrack et al., 2012).

Page 26257, Line 20: Please refer Khosrawi et al. (2011).

Page 26257, Lines 23—24: Please give references for the MW dates (e.g. Dörnbrack et al. 2012 and Kuttippurath and Nikulin, 2012).

Page 26257, Lines 23—26 and **Page 26258, Lines 1—3:** There are no PV or geopotential maps here, so appropriate references are needed to guide the readers to analyse the meteorological situation.

Page 26258, Line 18: Please introduce the figure as "Figure 2 shows..." and then present the results.

Page 26261, Lines 17—18: Specify, "compared to HCl observations" of MLS.

Page 26262, Lines 1—2: If there is a bias in the ERA Interim data, please also include the latitude region, time period, and reference for that. Also, emphasize that you have used ERAI for your simulations.

Page 26262, Lines 14—15: Please make this sentence more general for the ACE-FTS measurements, as the instrument has been measuring many trace gases since its launch, not only for this winter.

Page 26262, Lines 22—23: If you haven't used the data, any reason to say it? Please delete the sentence.

Page 26263, Lines 3—4: You have shown the ozone loss (in ppmv). Therefore, writing ozone loss rate is not suitable here. Either you calculate the loss rate during the period or just write loss.

Page 26263, Lines 5—6: Please specify the date of vortex split so that the reader can relate its impact on the ozone loss here.

Page 26263, Line 8: In general, the maximum ozone loss altitude will be around 475 K in cold winters and around 550 K in warm winters in the Arctic (e.g. Manney et al. 2011, Kuttippurath et al. 2010, Singleton et al., 2005, Gross et al., 2005, etc). Note that your ozone loss estimate from the MLS measurements also shows the peak ozone loss altitude of around 550K, which is in accordance with the situation for a warm winter. However, in your model simulations the peak ozone loss altitude (600—650 K) is still higher than these altitude ranges. Please also check your tracer simulations to know the reason for this.

Page 26263, Line 15: Please delete the bracket part here. The decrease in ClOx doesn't mean that there is large NOx influx to deplete ozone in that proportion. Please also give appropriate references for NOx induced ozone loss here.

Page 26263, Line 16: Kuttippurath et al. (2010) show that the average contribution of partial column ozone loss above 550 K to the total column ozone loss is about 19(+/-7) DU. This

amount is small, but cannot be neglected compared to the range of ozone loss estimates for the warm Arctic winters (e.g Harris et al., 2010, Kuttippurath et al., 2010, WMO, 07, and Goutail et al., 2005). What is the column ozone loss above 600 K in your simulations here ? Is it 77 – 58 DU, as sated in **Line 23** herein ?

Page 26263, Lines 7, 19—20: Compare your ozone loss estimates with other published results for this winter (e.g. Kuttippurath et al., 2010).

Page 26263, Line 21: Is it total column or partial column ? If partial, write the column range. Please note that this is not the ozone loss on 30 March, but it is the cumulative ozone loss since the model initialisation date.

One important point to consider here in the ozone loss estimate is the initialisation offset. There is an obvious offset between ozone loss estimates based on the simulations and MLS measurements. The MLS based loss estimate doesn't show zero on the day one (1 December). This should be zero (i.e. the passive ozone tracer should be the same as the actual ozone) and should be corrected for the whole period. Please check your modelled and measured loss estimates and correct accordingly. You could also smooth the time series for a few days.

Page 26263, Line 23: Please specify the column range (e.g. 350—600 K) for the estimates of 77 DU and 58 DU and compare with other available loss estimates for the same column ranges and time period.

Page 26263, Lines 23—25: Do you mean that the ozone loss estimated by the passive technique (that you used here) is more comparable to the loss estimated by the vortex averaged method and they do not yield significant column ozone loss above 600 K? It is not a good idea to state that without extensive comparisons of the loss estimates using all other techniques for the same winter under same criteria. In addition, you need to compare partial column and total column estimates to arrive into such a conclusion. Therefore, please rewrite these sentences.

Page 26263, Line 27: Please give a range of ozone loss estimates (from very cold winter estimates to very warm winters) and then compare your estimates to others to state whether the derived ones are “moderate” or not.

Page 26263, Line 28: If you mean Figure 4 of Manney et al., then these are the ozone loss estimates in mixing ratio with altitude and not the column ozone loss. You could refer Harris et al. (2010) or WMO (2007) for this purpose. There are also other references (Tilmes et al. 2006, Goutail et al. 2005, etc.) for the long-term column ozone loss estimates for comparisons.

Page 26264, Lines 10—13: Did you consider your vortex definition at each model level ? Please also state this in an appropriate place, preferably in Section 3.2, where you start the discussion of the vortex-averaged ClO analyses.

Page 26265, Paragraphs 1—2: Please refer and compare the results with Khosrawi et al. (2011) here. Also, it is necessary to compare with similar denitrification studies for the past winters (e.g. Feng et al. 2011, Kleinboehl et al., 2005, Grooss et al., 2005, etc).

Page 26266, Lines 5—8: "In addition, it excludes". Could you please make this sentence more clear ?

Page 26270, Line 7: This has been stated more than 3 times in this manuscript. You could either restrict your analysis up to February or delete this statement, as it indicates that there is a lot of mixing and no vortex boundary.

Page 26271, Lines 16—18: The timing and altitude dependence of chlorine activation did not affect ozone simulations ? Perhaps, I did not understand this point.

Page 26271, Lines 19—20: They have already published their results. Therefore, your results confirm their findings, not the reverse ?

Table 3: Please write in the caption that you list the sensitivity runs with respect to the reference run (e.g. MINUS-ONE-KELVIN : Global temperature offset of -1 K with respect to the reference run).

Figure 1: Line 5, replaced the constant value at all model levels ? If yes, please state that here.
Lines 5—6: What I understood is that the white lines (infact, they are CONTOURS) actually show the possible area (altitude versus time) of NAT and ice clouds, not the time periods.

Figure 2: ClO plots: Why these ClO values are very small? I would expect the maximum values of around 1.8 ppbv in both simulations and measurements (e.g. Manney et al. 2011, Kuttippurath et al. 2010, Santee et al. 2008, etc). Here the maximum is just 1 ppbv. Your other ClO comparisons too show peak values of around 1.6 ppbv. Is it possible to select the matching profiles with respect to time and SZA ? Otherwise, I would suggest you to remove this figure to avoid misunderstandings of the readers (you also state that the quantitative comparison is meaningless). You can still discuss your results with the ClOx and other ClO comparison plots. What about the vortex criteria? In Figure 1 you have used two criteria. Could you please make the legends and labels slightly larger for this figure ?

Figure 2: HNO3 plots: There are white patches (missing values) in MLS HNO3 in the upper stratosphere. Therefore, the difference (model simulations – MLS measurements) should also show the white patches for the same period. But I do not see them there. This is also the same for the N2O plots!

Figure 3: ClO plots: Averaged over a particular altitude band or just at 46 hPa?
Please put a colour bar also for the difference (on the right).

Figures 4, 5, 6, 7, 9, 10 and **12:** Delete the "Colors indicate... runs" and write something like "Results from various sensitivity runs are given in respective colors".

Figure 8: "as a function of time and potential temperature." **Line 2**: What you have shown is the reverse i.e. "ozone-passive ozone tracer" (see the negative ozone loss scale). Please change the sentence. The white contours show the possible area of NAT and ice clouds, not the time periods.

Figure 9: This is the cumulative ozone loss on 30 March, not the ozone loss on 30 March. Also shown is the ozone – passive ozone tracer, not as stated here. Please correct the initialisation error for the ozone loss estimate based on MLS observations (if there is).

Figure 10: "as a function of time". Correct the initialisation offset of MLS based ozone loss estimate. Please change that the shown curves are ozone-passive ozone tracer not the reverse as stated in the caption.

Figure 11: "as a function of time and ...". Please also write what the white contours are (NAT and ice cloud area).

TECHNICAL CORRECTIONS

Page 26247, Line 1: It indicates that there is no chemistry in denitrification.
Page 26247, Line 10: Please define the abbreviations MLS and ACE-FTS
Page 26249, Line 7: Please define the abbreviation MLS
Page 26249, Line 8: Please define the abbreviations ACE-FTS and SCISAT
Page 26249, Line 11: "in the framework of "
Page 26249, Line 13: "In a related study" is enough
Page 26249, Line 17: Delete "Polar Stratospheric Clouds". You have already spelled this out.
Page 26251, Line 15: Please define the abbreviation SAGE
Page 26252, Line 5: "Carslaw et al. (2002). Further details about the scheme can be found ..."
Page 26252, Line 11: "enough to be neglected"
Page 26254, Line 11: "but in no heterogeneous" ?
Page 26254, Line 26: Please start the sentence properly, not with the E.g. acronym.
Page 26256, Line 3: Please define the abbreviation HALOE
Page 26257, Line 4: Please define CFCs. Also in the same line "significant" or "relevant" ?
Page 26257, Line 13: "early December 2009"
Page 26257, Line 21: "Starting a few days later..". This sentence is not clear. Please rephrase it.
Page 26263, Line 3: Replace " highest " by "largest"
Page 26263, Lines 17—18: " the results presented for HCl and ClO in Section 3.3."
Page 26263, Line 28: "The column ozone loss"
Page 26265, Line 21: Change to vortex-averaged data (not observations).
Page 26265, Line 24: compatible or comparable with ?
Page 26266, Line 7: "during the GEOPHYSICA flights" ?
Page 26266, Line 10: Replace additionally with "also".

Page 26266, Lines 11—12: “The tracer measurements from the flights show...”
Page 26266, Line 18: “mixing between model and reality” ?
Page 26266, Lines 19—22: Please start with “For instance or For example”, not with E.g.
Page 26267, Line 3: “was observed in 2009/2010”
Page 26267, Line 14: Replace “is not possible” by “is difficult”.
Page 26267: Lines 23—29: Is this just one sentence ? If yes, please split this.
Page 26267: Lines 25—26: “relatively smaller than the ABBATT” rates .
Page 26268: Para 1 and 2: Merge both of these.
Page 26268: Line 23: Delete “too”
Page 26269: Line 2: Please start the sentence with a proper word (not with E.g.)
Page 26269: Line 15: Write something like, “. Note that there is a persistent bias in our simulations...”. Otherwise, this sentence is too negative now.
Page 26269: Line 17: Delete “we believe that”
Page 26269: Line 18: “those are less affected by ” ?
Page 26269: Line 24: Delete “Fortunately”
Page 26278, Line 1: Replace this with their ACPD article.
Page 26278, Line 16: Replace with their ACP article.

REFERENCES:

Dörnbrack, A., Pitts, M. C., Poole, L. R., Orsolini, Y. J., Nishii, K., and Nakamura, H.: The 2009–2010 Arctic stratospheric winter – general evolution, mountain waves and predictability of an operational weather forecast model, Atmos. Chem. Phys., 12, 3659–3675, doi:10.5194/acp-12-3659-2012, 2012.

Drdla, K. and Müller, R.: Temperature thresholds for chlorine activation and ozone loss in the polar stratosphere, Ann. Geophys., 30, 1055-1073, doi:10.5194/angeo-30-1055-2012, 2012.

Feng, W., Chipperfield, M. P., Davies, S., Mann, G. W., Carslaw, K. S., Dhomse, S., Harvey, L., Randall, C., and Santee, M. L.: Modelling the effect of denitrification on polar ozone depletion for Arctic winter 2004/2005, Atmos. Chem. Phys., 11, 6559-6573, doi:10.5194/acp-11-6559-2011, 2011.

Grooß, J.-U., Günther, G., Müller, R., Konopka, P., Bausch, S., Schlager, H., Voigt, C., Volk, C.M., and Toon, G. C.: Simulation of denitrification and ozone loss for the Arctic winter 2002/2003, Atmos. Chem. Phys., 5, 1437-1448, doi:10.5194/acp-5-1437-2005, 2005.

Khosrawi, F., Urban, J., Pitts, M. C., Voelger, P., Achtert, P., Kaphlanov, M., Santee, M. L., Manney, G. L., Murtagh, D., and Fricke, K.-H.: Denitrification and polar stratospheric cloud formation during the Arctic winter 2009/2010, Atmos. Chem. Phys., 11, 8471–8487, http://dx.doi.org/10.5194/acp-11-8471-2011doi:10.5194/acp-11-8471-2011, 2011.

Kleinboehl, A., Bremer, H., von Koenig, M., Kuellmann, H., Kuenzi, K., Goede, A. P. H., Browell, E. V., Grant, W. B., Toon, G. C., Blumenstock, T., Galle, B., Sinnhuber, B. M., and Davies, S., Vortex-wide denitrification of the Arctic polar stratosphere in winter 1999/2000 determined by remote observations, J. Geophys. Res., 107, 8305, doi:10.1029/2001JD001042, 2002.

Kleinboehl A., Bremer, H., Kuellmann, H., Kuttippurath, J., Browell, E. V., Canty, T., Salawitch, R. J., Toon, G. C., and Notholt, J.: Denitrification in the Arctic mid-winter 2004/2005 observed by Airborne SUbmillimeter Radiometry, Geophys. Res. Lett., 32, L19811, doi:10.1029/2005GL023408, 2005.

Kuttippurath, J. and Nikulin, G.: A comparative study of the major sudden stratospheric warmings in the Arctic winters 2003/2004–2009/2010, Atmos. Chem. Phys., 12, 8115-8129, doi:10.5194/acp-12-8115-2012, 2012.

Kuttippurath, J., Godin-Beekmann, S., Lefèvre, F., and Goutail, F.: Spatial, temporal, and vertical variability of polar stratospheric ozone loss in the Arctic winters 2004/2005–2009/2010, Atmos. Chem. Phys., 10, 9915-9930, doi:10.5194/acp-10-9915-2010, 2010.

Newman, P. A., et al., An overview of the SOLVE-THESEO 2000 campaign, *J. Geophys. Res., 107*(D20), 8259, doi:10.1029/2001JD001303, 2002.

Santee, M. L., Lambert, A., Read, W. G., Livesey, N. J., Manney, G. L., Cofield, R. E., Cuddy, D. T., Daffer, W. H., Drouin, B. J., Froidevaux, L., Fuller, R .A., Jarnot, R. F., Knosp, B. W., Perun, V. S., Snyder, W. V., Stek, P. C., Thurstans, R. P., Wagner, P. A., Waters, J. W., Connor, B., Urban, J., Murtagh, D., Ricaud, P., Barrett, B., Kleinböhl, A., Kuttippurath, J., Küllmann, H., von Hobe, M., Toon, G. C., and Stachnik, R. A.: Validation of the Aura Microwave Limb Sounder ClO measurements, J. Geophys. Res., 113, D15S22, doi:10.1029/2007JD008762, 2008.

Singleton, C. S., Randall, C. E., Chipperfield, M. P., Davies, S., Feng, W., Bevilacqua, R. M., Hoppel, K. W., Fromm, M. D., Manney, G. L., and Harvey, V. L.: 2002-2003 Arctic ozone loss deduced from POAM III satellite observations and the SLIMCAT chemical transport model, Atmos. Chem. Phys., 5, 597-609, doi:10.5194/acp-5-597-2005, 2005.

Tilmes, S., Müller, R., Engel, A., Rex, M., and Russell III, J. M.: Chemical ozone loss in the Arctic and Antarctic stratosphere between 1992 and 2005, *Geophys. Res. Lett.*, 33, L20812, doi:10.1029/2006GL026925, 2006.

Wegner, T., Grooß, J.-U., von Hobe, M., Stroh, F., Sumiñska-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.

WMO (World Meteorological Organisation): Scientific assessment of ozone depletion: 2006, Global Ozone Research and Monitoring Project-Report No 50, 572~pp., Geneva, Switzerland, 2007.