

Interactive comment on “Uncertainties in modeling heterogeneous chemistry and Arctic ozone depletion in the winter 2009/2010” by I. Wohltmann et al.

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Dear reviewer,
thank you for reviewing our paper and your very helpful comments!

Main points

- 1) We have now included the uncertainty ranges for column ozone loss and chlorine activation in the abstract. Some more conclusions from the various sensitivity experiments were included in the abstract, in a similar manner as in the conclusions. We also added a bullet to the conclusions (“Current estimates. . .”). I would

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like to keep the abstract relatively short, since in my personal experience, I tend not to read papers with a long and complicated abstract.

- 2) We have added a section about the satellite instruments. Discussion and a table have been added on validation, statistical and systematic errors and altitude resolution. Statistical error bars have been added to Figure 7. Information on the vertical coverage has been omitted, since the vertical ranges of the measurements are usually greater than the vertical range of the model. The horizontal coverage is only mentioned for ACE, where it is limited. Horizontal and vertical resolution of the satellite measurements are comparable to the horizontal and vertical resolution of the model, so no further measures are taken when comparing the measurement data to the model results.
- 3) The main reason is that the RECONCILE measurement campaign took place in this winter and that there are more datasets to compare with than in other winters. We have now added “This winter was selected due to the large amount of different data sets available from the campaign for comparison in this winter.” to the introduction. There was no special meteorological reason to choose this winter.
- 4) The meteorological situation is needed as background information for the paper, but is no central issue. We would like to keep this section short. At the beginning of the section, the reader is pointed to the paper of Dörnbrack et al., in which a comprehensive overview over the dynamical situation in the winter 2009/2010 is already available. We think this is sufficient and that more information than given in Section 3.1 is not needed to be able to understand the following discussion. The description is also easy to understand without additional figures. See also our reply to the specific comments below.
- 5) It is not clear to us if you mean comparison to measurements or comparison to other models here. Ozone loss is a special case since it is a combination of mea-

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surement and modeling and there are several fundamentally different methods of deducing ozone loss. We will reply to all possible interpretations:

- a) Comparison to measurements: Model results are extensively compared to measurements, both in the paper and in the supplement. With a few exceptions, all data available for this winter have been used, including satellites, ozone sondes and several aircraft instruments. There are only few measurements left which we did not use and which would not add much additional insight. We think it would also not add much insight to the paper to compare with measurement results shown in other papers. We put much effort in obtaining meaningful comparisons (e.g. averaging, vortex definition, box model runs for the shorter lived species) and comparing just to some figures or values from other papers would be inferior to that.

Regarding the ClO comparison: There is a paper by Suminska-Ebersoldt et al. which is cited in the paper. The measurements discussed in Suminska-Ebersoldt et al. are compared with the model results in the supplement. We are not aware of any additional measurements apart from the MLS and Geophysica measurements, which are compared to the model in the paper.

- b) Comparison to other models: This is certainly interesting and has some justification, but is beyond the scope of the paper. The important point for our paper and the most important measure for model performance is how well the model results compare to reality, i.e. to measurements. We acknowledge that there can be interesting insights when comparing two models to each other (e.g. if two different concepts in the heterogeneous chemistry are realized or if you compare conserved tracers in Lagrangian and Eulerian models), but we think that is of secondary importance for this paper. This is no model validation or model comparison paper. Citing just a few random results from other models would not add much insight without a detailed interpretation. In addition, this is already a 20 page paper plus sup-

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plement. We think a comparison to other models is better suited to appear in a separate study.

- c) Comparison of ozone loss estimates: This is certainly justified, since different methods of calculating ozone loss can yield quite different results and one needs to take care to compare to the right quantity in the model. Thanks for pointing us to the Kuttippurath paper, which we refer to now. In the moment, we are not aware of any other study giving ozone loss estimates for the 2009/2010 winter.

Unfortunately, there seems to be a shortcoming in the calculation of the ozone loss estimates in Kuttippurath et al. (see the specific comments section below, page 26263, line 7, 19–20). We did not add discussion on this issue and the differences between the results from our study and Kuttippurath et al., since we think that would distract the reader.

- d) Denitrification: We refer to the Khosrawi et al. study now. However, while it contains the word “denitrification” in the title, denitrification is not modelled in this study (see the specific comment section, page 26265, paragraphs 1–2). We are not aware of any other studies modelling denitrification for this winter.

Specific comments

- Page 26247, lines 11–14: The results in the paper show that the relative differences in column ozone between the sensitivity runs are small, while the differences in other species are larger (see e.g. Fig. 10 for the column loss or Fig. 4 for ClOx). We added “column” in the abstract.
- Page 26247, lines 14–17: We are a little bit hesitant to write a recommendation in the abstract or to give further implications. The role of liquid vs. solid PSCs in ozone depletion is a complicated issue. Not a small number of people tend only

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to read the abstract and conclusions of a paper. We would prefer that people actually read the paper before they come to conclusions that are problematic or change their models without really understanding what has been written.

- Page 26248, lines 1, 9: The relevant publications are cited in line 6.
- Page 26248, line 10: We have now cited some of the papers a second time in the introduction, which we originally cited on page 26253, line 5–14.
- Page 26248, line 22: Done.
- Page 26249, line 1: We have added a reference for the SOLVE/THESEO overview paper and a reference to a VINTERSOL-EUPLEX paper.
We don't think that the abbreviations should be expanded. It does not add any insight to the text to know that SOLVE/THESEO means "Stratospheric Aerosol and Gas Experiment III Ozone Loss and Validation Experiment and the Third European Stratospheric Experiment on Ozone". It does clutter the text and distracts the reader.
- Page 26249, line 17: Added "depletion of gas-phase HNO₃".
- Page 26249–26250: Done.
- Page 26250, line 5: It is correct that there is no numerical diffusion in purely Lagrangian models. The mixing ratio of a species on a trajectory does not change by a change of the position of the trajectory and if there is no interaction between different air parcels.
- Page 26250, lines 13–15: Rephrased the sentence as suggested.
- Page 26253, lines 11–14: We changed "was" to "is".

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- Page 26253, lines 21–22: Paragraphs merged. Added year to the citation. What do you mean by "introduce Table 3 properly"? Seems fine to us.
- Page 26254, lines 25–28: Done.
- Page 26255, lines 17–19: We think it is ok to write "show". Obviously, this sentence was written after looking at the results. But we see no problem with anticipating the results here.
- Page 26257, line 15: We added Kuttippurath and Nikulin as reference. Note that we did not calculate the amplitude of wave 2 here. The existence of a wave 2 event was derived from looking at maps of the polar vortex.
- Page 26257, line 20: The citation seems not really obvious to us (the paper is about denitrification mainly and not about the meteorological evolution). In addition, this is a simple statement which is easily derived from ECMWF data, so we think a citation is not really needed.
- Page 26257, lines 23–24 and Page 26257, lines 23–26 and page 26258, lines 1–3: We changed the formulation of the first paragraph to make it clearer that the reader is supposed to refer to the Dörnbrack et al. paper in case he wants to get a more detailed overview. We think it is sufficient to cite this paper only once in this relatively short section. In addition, we think the description is easy to understand without PV maps. The meteorological situation is needed as background information for the paper, but is no central issue.
- Page 26258, line 18: We rephrased the sentence as suggested.
- Page 26261, lines 17–18: Done.
- Page 26262, lines 1–2: We rephrased the sentence. We do not claim that there actually is a bias in the ECMWF data. This is about possible reasons for the discrepancy and the bias is just proposed as one of several possibilities.

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- Page 26262, lines 14–15: We have rephrased the sentence.
- Page 26262, lines 22–23: Deleted.
- Page 26263, lines 3–4: We rephrased the sentence.
- Page 26263, lines 5–6: Added “in early February”. The exact date is given in the discussion paper in Section 3.1, page 26257, line 26.
- Page 26263, line 8: You are right, there is an obvious discrepancy between the ozone loss estimates from the model and from MLS between 500 K and 650 K. Note that these estimates are not independent of each other, since both use the same passive profile. It is the same to compare ozone between model and MLS. We have now added some discussion on the reasons of the discrepancy to the paper. The life time of ozone between 500 K and 650 K at the end of March is of the order of months, so the discrepancy can be caused by chemistry on longer time scales or by transport effects. Correspondingly, there are two plausible explanations for the discrepancy in ozone:
 - a) The discrepancy could be caused by differences in dynamics between model and reality. There is a corresponding discrepancy between modelled and measured N₂O, N₂O is overestimated in the model. The discrepancies in ozone and N₂O appear when the major warming sets in at the end of January. They could be explained if there was more mixing over the vortex edge in the model (i.e. in ERA Interim) compared to reality during the major warming. The gradients of N₂O and ozone over the vortex edge in this altitude region would be compatible with that. Differences in subsidence can be excluded, since more ozone in the model would imply more subsidence in the model than in reality. However, that would mean less N₂O in the model, and not more N₂O, as actually modelled.
 - b) Additionally, part of the discrepancy in ozone could be caused by chemistry: There is an overestimation of HCl and an underestimation of active chlorine in

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the model in January just in the altitude range where the discrepancy in ozone appears. That does not explain the discrepancy in N₂O, however.

- Page 26263, line 15: We have added Konopka et al. (2007) as a reference. We have added “and the increase in NO_x in Fig. S18 of the supplement” to the sentence to be more clear. We will not delete the bracket part. While it is true that the decrease in ClO_x does not necessarily imply that NO_x increases, it implies that ozone loss cannot be caused by ClO_x.
- Page 26263, line 16: We do not claim that it can be neglected and just say it is “relatively” small. You are right, the column loss above 600 K is 77-58=19 DU. We think that can be deduced easily from the text and no further clarification is needed.
- Page 26263, lines 7, 19–20: Thanks for pointing us to the Kuttippurath paper. In the moment, we are not aware of any other paper giving ozone loss estimates for the 2009/2010 winter.

We did comparisons with Kuttippurath et al. (2010) for the end of February and the method with the passive tracer and MLS. At 475 K, both values agree well (both are 0.9 ppm), but at 675 K, there is a large discrepancy (1.1 ppm in our paper, estimated less than 0.5 ppm in Kuttippurath et al.).

In our opinion, there is a shortcoming in the calculation of the ozone loss values in Kuttippurath et al. (2010), at least at 675 K and higher. 475 K may not be affected. As in our approach, a passive ozone profile is used to calculate ozone loss by subtraction of this profile from MLS data. It is important for the passive profile where the upper boundary of the model is located. Air is transported downward in the polar vortex in the course of the winter and will enter through the upper model boundary. At the upper model boundary chemical species and passive tracers have to be initialized when they enter the model domain from above. However, this is not possible for the passive ozone profile (at least not

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with meaningful values), since it is not known where the air was situated on the day when the passive tracer was initialized. Therefore, it is necessary to restrict the analysis to altitudes, where the air masses originate from below the upper model boundary (on the day of initialization of the passive tracer). With an upper boundary of 1900 K and initialization on 1 December, this is at about 750 K at the end of the winter (that is the reason to cut most plots at 750 K). If we understand it correctly, the upper boundary of the Mimosa-Chim model is at 950 K (the value is from Tripathi et al., 2006). This probably means that values of the passive ozone profile and consequently ozone loss at 675 K are not reliable. This is supported by the fact that there is no ozone depletion visible above 675 K for 2010 and MLS data, which does not seem to be realistic.

For this reason, we are a little bit hesitant to discuss the discrepancy in the paper in the same way as done above. We are of the opinion that would distract the reader.

The modelled ozone loss of Mimosa-Chim and of ATLAS compare surprisingly well at the end of February (about 0.9 ppm in a large altitude range), but the results for Mimosa-Chim will also be affected by the problem with the passive profile.

- Page 26263, line 21: The plot shows the total column of the model. We have clarified that in the text. We have corrected for the offset in the plot and changed the value for the MLS column loss in the text accordingly.

The offset between the ozone loss estimate from observations and the model is caused by differences in averaging. The passive ozone is averaged over all air parcels of the model inside the vortex, with the passive ozone interpolated from the position of the satellite measurements to the position of the air parcels in the first model time step. The MLS ozone is averaged directly over all satellite measurements inside the vortex.

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- Page 26263, line 23: We have added “350–1900 K” for the complete column. Comparison to other estimates: See the comment above (line 7, 19–20).
- Page 26263, lines 23–25: We think there is a misunderstanding here, this is not what we wanted to express. We have rephrased the paragraph to avoid confusion. We just wanted to express that the results of the vortex average method (in Rex et al.), which are given for the partial column below 550 K, should be compared to the results of the passive tracer method (using the results of our model) by taking the partial column below 550 K in our model. In addition, we erroneously used 600 K instead of 550 K, which has been corrected.

In particular, we don't want to imply that the passive tracer method does not yield significant ozone loss above 550 K.

The vortex average method, as applied for example by Rex et al. (2006) with ozone sondes, typically gives no reliable results above about 550 K. The reason for this is that a vortex averaged passive ozone profile is needed in the method, which gives no reliable results above 550 K (due to transport over the vortex edge in higher altitudes and the upper boundary of the profile descending down in the course of the winter).

It has been added to the text that the partial column of the vortex average method in Rex et al. is 380–550 K. See also our reply to your comment on Page 26263, line 28.

- Page 26263, line 27: The range of ozone loss estimates was given in the text: The upper limit was given in the sentence, together with a reference. The lower limit (20 DU) was missing, since it was implicitly assumed that it was near 0 DU and has been added to the text now. The results were compared to Rex et al. as a comparison to others in the text, as requested. We think that it is sufficient to cite only this study here, since it encompasses all years for which ozone loss estimates based on measurements are available (if including the unpublished

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update mentioned in the text), and all other studies known to us do include only a subset of these years. The statement of a moderate loss is based on the comparison of 43 DU to 130 DU given in the text.

- Page 26263, line 28: Including the citation of Manney et al. at the position where it was was misleading. We have rephrased the text to be more concise here: We have added that the results are results produced by us, added the column range (380–550 K) and moved the citation of the Manney paper into a separate sentence.

The range of up to 130 DU given in the text is based on results of the vortex average method and is based on our own work and sonde data from the Match campaigns coordinated from our institute. The most recent publication showing an overview of these values is Rex et al., 2006, and some of the values are still unpublished, including the winter 2010/2011, which showed the record ozone loss of 130 DU.

The value is not based on Figure 4 of Manney et al., and it is also not based on Figure 5 of Manney et al. (which shows values of up to 130 DU, but the figure has been produced with a simplified method not applicable here).

We think it is sufficient to cite only the Rex et al. study here (see last comment above for justification). The references that you propose don't include the winter 2010/2011, which is the maximum of the time series. A comparison of the ozone loss in different winters is clearly beyond the scope of our case study for the winter 2009/2010.

- Page 26264, lines 10–13: The vortex definition was derived from the Nash criterion at 475 K, then transformed to equivalent latitude and applied at all altitude levels. This information was missing in the paper and has been added to the caption of Figure 1.

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- Page 26265, paragraphs 1–2: We refer shortly to the Khosrawi paper at the beginning of Section 3.5 now. The modelling study in Khosrawi et al. is a case study for a few trajectories and only models liquid STS droplets and does not model the denitrification, so it is not possible to compare anything to the modelled denitrification of our model. Comparison to the HNO₃ measurement data in Khosrawi et al. gives no new insights: The MLS data is the same as in our study, and the Odin/SMR data is very similar to the MLS data.

We would like to do without the discussion of denitrification studies for past winters. It is beyond the scope of the paper (which is a case study for the winter 2009/2010) to model other winters or to compare to other winters.

- Page 26266, lines 5–8: We removed “In addition” from the sentence.
- Page 26270, line 7: We rephrased the sentence.
- Page 26271, lines 16–18: We rephrased the sentence.
- Page 26271, lines 19–20: Changed the sentence.
- Table 3: Done.
- Figure 1, line 5: Done.
- Figure 1, lines 5–6: Replaced “lines” by “contours”. “Altitude range” added.
- Figure 2, CIO plots: The most accurate way to select matching profiles is to apply the method used in Figure 3 of the discussions manuscript. We did not apply this method in Figure 2 of the discussions manuscript, because it required a large number of box model and trajectory runs (several million runs, which add up to several weeks of computing time). We have now decided to recalculate the data in Figure 2 with the method used in Figure 3. We have done the necessary changes in the text and the caption. Note that the order of the figures has

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changed, since the former Figure 2 is now mentioned for the first time later in the text. Figure 2 is now Figure 3 and vice versa. The bias in CIO is still existent in the new figure.

The sampling of the vortex by the satellite data and the model data is quite different: While the vortex is sampled quite uniformly in the model (both vertically and horizontally) at a fixed time, the vortex is sampled at different times in the MLS data, only at discrete pressure levels and with decreasing horizontal density in lower latitudes. That means that there is a trade-off when calculating vortex averages: Either the best estimate of the vortex mean can be obtained (sample at the model locations and time) or the best comparison of model and MLS (sample at the satellite locations and times). We have opted for the latter here now.

- Figure 2, CIO plots, “What about the vortex criteria?”: Do you mean the Nash criterion vs. the fixed value? This is applied at different times, so there is no ambiguity.
- Figure 2, CIO plots, “Could you please...”: Font size has been increased as suggested.
- Figure 2, HNO₃ plots: The white patches are either missing values or negative values in the MLS data. Added “Negative values and missing values are shown in white” to the caption. We don’t want to assign a color to the negative values. This would force us to change the color bar to include negative values both in the left column for the model (where we have no negative values) and in the middle column. That would be more confusing than the way it is done in the moment. Additionally, we have corrected the range for the N₂O difference.
- Figure 3, CIO plots: The MLS data are available at several retrieval levels. The data on the levels are an average over a certain altitude range. This is a property of the retrieval algorithm. That is the reason why the caption says “MLS layer

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centered at 46 hPa”. The box model runs end exactly at 46 hPa, but their initialization requires vertical interpolation and thus depends on the vertical model resolution. Added the missing colour bar.

- Figures 4, 5, 6, 7, 9, 10, 12: Changed to “The different sensitivity runs are represented by different colors”.
- Figure 8: Changed to “as a function of time”. Exchanged “ozone” and “passive tracer”. Time periods: see above.
- Figure 9: Exchanged “ozone” and “passive tracer”.
- Figure 10: Changed to “as a function of time”. Added offset to initialization. Exchanged “ozone” and “passive tracer”.
- Figure 11: Changed to “as a function of time”. Added explanation of contours.

Technical corrections

- Page 26247, line 1: We wanted to express that there is also a microphysical component in denitrification.
- Page 26247, line 10: The abbreviations are defined in the new section about the satellite instruments.
- Page 26249, line 7: See above.
- Page 26249, line 8: See above.
- Page 26249, line 11: Done.
- Page 26249, line 13: Done.

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- Page 26249, line 17: Done.
- Page 26251, line 15: Is now defined in the new section about satellite instruments.
- Page 26252, line 5: Done.
- Page 26252, line 11: Done.
- Page 26254, line 11: Done.
- Page 26254, line 26: Changed to “For example”.
- Page 26256, line 3: Is now defined in the new section about satellite instruments.
- Page 26257, line 4: Expanded “CFCs”. Replaced “relevant” by “significant”.
- Page 26257, line 13: Done.
- Page 26257, line 21: Done.
- Page 26263, line 3: Done.
- Page 26263, lines 17–18: We assume you mean page 26264. Done.
- Page 26263, line 28: Done.
- Page 26265, line 21: Done.
- Page 26265, line 24: “compatible” is correct.
- Page 26266, line 7: Done.
- Page 26266, line 10: Done.

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- Page 26266, lines 11–12: Done.
- Page 26266, line 18: Exchanging “mixing” and “heating rates” should remove the ambiguity.
- Page 26266, line 19–22: Changed to “For example”.
- Page 26267, line 3: Done.
- Page 26267, line 14: Done.
- Page 26267, lines 23–29: Shortened.
- Page 26267, lines 25–26: That would not have the same meaning.
- Page 26268, paragraphs 1 and 2: Would like to keep that to emphasize the difference between the rates on liquid aerosol and NAT.
- Page 26268, line 23: Done.
- Page 26269, line 2: Changed to “For example”.
- Page 26269, line 15: Done.
- Page 26269, line 17: Done.
- Page 26269, line 18: Done.
- Page 26269, line 24: Done.
- Page 26278, line 1: Done.
- Page 26278, line 16: Done.