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Comment

***Interactive comment on “Impact of energetic particle precipitation on stratospheric polar constituents: an assessment using MIPAS data monitoring and assimilation” by A. Robichaud et al.***

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Interactive comment on “Impact of energetic particle precipitation on stratospheric polar constituents: an assessment using MIPAS data monitoring and assimilation” by A. Robichaud et al.

Reply to referee 1

The authors would like to thank Referee 1 for his/her thoughtful review and among other things for pointing out some relevant publications which in the end have contributed to

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make this document more comprehensive and have helped to clarify the context of this study.

Referee 1: Major suggestions for improvements:

1. Unfortunately you do not discuss the study by Vogel et al. (ACP, 2008), who used MIPAS observations of NO<sub>x</sub> as an upper boundary condition for their model calculations with CLAMS. Therefore, in several aspects this study is similar to yours and should be discussed/compared to.

Reply:

We appreciate the comment and have included this reference with a short discussion in the revised version. However, it is important to note that our modeling and assimilation systems differ from that of Vogel et al., especially concerning the presence of some EPP modeling in the latter (as a form of an upper boundary condition), and the presence of full assimilation capabilities with the 3D-VAR FGAT (variational assimilation with first guess at appropriate time) for MIPAS temperature and chemical constituents in the former. Moreover, the transport, the way and the frequency to which data initialization are performed are also different. This would render a comparison a rather difficult and tedious task since the basis of both systems is substantially different. We also believe that such task is not within the focus of our paper. Nevertheless, in the revised version, we agree to compare the final results, that is the magnitude of ozone loss obtained with two other existing and well established methods to study EPP: i.e. 1) comprehensive modeling of EPP with neutral and ion chemistry (for example, Jackman et al., 2005) and 2) the method of imposing an upper boundary condition of NO<sub>x</sub> simulating EPP as in Vogel et al. (2008). Therefore, following the reviewer's recommendation, reference to the Vogel et al. paper and a brief discussion of the two other approaches and results concerning the impact on total ozone column for the SPE of October- November 2003 period have been added in the revised version (see also details on the reply of further comment no. 7 below). This is done in section 5.4 of

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the manuscript dealing with the evaluation of polar loss for the “Halloween” storm. We think that the method of OmF 2-D time series shown in our paper (Figs. 2-5 and 7-9) to study the impacts of EPP is original and could be viewed as a complementary method to existing approaches to study EPP. We also wish to add here and emphasize that our contribution in this study goes beyond that of the analysis of the impact of ozone loss in the polar vortex, but also addresses several aspects related to the data assimilation methodology itself and derived diagnostics, more specifically: 1) the demonstration on both theoretical and experimental basis that our assimilation system almost eliminates the chemical biases and reduces the random error in the case of slow time scale model errors or imprecise initial conditions while having less impact on fast time scale model errors. Note that EPP phenomena are interesting in this context because, among other things, they produce impacts of various time scales, 2) showing that the chemical assimilation of MIPAS-ESA in the context of EPP is successful if careful attention is paid to proper error statistics and quality control, 3) presentation of a new diagnostic tool (made up of 4 panels) to study the impact of phenomena producing large effects which a priori are not necessarily present in the modeling system and perform an analysis with the OmF technique,

A mention of the above 3 points has been added in the introduction of the revised version in order to clarify the context of our study.

Referee 1:

2. The OmF technique obviously shows all differences between model and observations, including those not related to EPP. Maybe using the same technique for the year 2004, with much less EPP, could show how well this chemical assimilation scheme is performing? Additional features like the HNO<sub>3</sub> problem mentioned at 22475, l18 could then possibly be identified.

Reply:

In the experiments that we have conducted, the effect of EPP is not always prominent

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and significant at all levels (e.g. below 20 hPa) and the diagnostic on those time periods and regions where the EPP effect is much reduced already provide evidence of the validity of the OmF diagnostic. We do not feel that it is necessary to include another year of assimilation in this paper to prove this point. In fact, both modeling and assimilation systems have been fully validated using independent data in few previous studies (using HALOE, ozonesonde data, GOMOS, TOMS for chemical constituents and temperature, see Ménard et al., 2007; de Grandpré et al., 2009) and from ground-based FTS measurements for HNO<sub>3</sub> (Batchelor et al. 2009). The periods of verification for meteorology and chemistry were: Aug-Dec 2003 (a strong EPP period) and Feb-March 2003 (a moderate EPP period). In the case of HNO<sub>3</sub>, the verification period was 2007 (a weak EPP period). Generally speaking, the verification has shown good to excellent agreement in the stratosphere for O<sub>3</sub> and HNO<sub>3</sub> for cases of weak to moderate EPP effects. Therefore, concerning that matter, we are confident with the results from our model and assimilation system not only for the period used in the paper but also for other years. Now, one of the mismatch revealed by the verification process (aside from the huge EPP signature for NO<sub>2</sub> and HNO<sub>3</sub> during strong EPP periods) appears on figure 3a below 10 hPa (before day 280). This residual has been associated to a misrepresentation of the process of removal of HNO<sub>3</sub> during denitrification giving strong negative OmF in our model. It slowly disappears after the end of the polar night and is not present during the following summer in the Antarctic region. On the other hand, in the Arctic region (figure 8a), another mismatch shows up that is a slow build up of the HNO<sub>3</sub> OmF around 25 hPa (labeled as 4 on the figure) which is clearly not an effect of EPP (since the latter has started before the onset of the SPE which took place at the end of October around day 301). Notice that, with assimilation (both in Figs. 3c or 8c), the HNO<sub>3</sub> problems are almost eliminated, indicating slow time scale errors (in the case of Fig. 8c), imprecise initial conditions or both (in the case of Fig. 3c). This issue had already been identified in Ménard et al. 2007 and reported at 22475, I18 (as noted by referee 1). We have re-worded the sentence to make it clearer and put emphasis that these problems are not related to EPP since they are also present in other

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non-EPP period (i.e. during 2007). The nature of these biases deserves further investigation but we believe that they do not influence the quality of the current study. On the contrary, one of the outcomes of the presented in our paper is in having identified a method that can be used to precisely locate and help diagnosing misrepresentation problems by following their spatio-temporal evolution through the OmF technique.

Referee 1

3. A major concern is the lack of comparison to the MIPAS-IMK products or publications, which have been shown to be very well suited to study these events. This is briefly mentioned in section 2.3, and at 22477 I16 but in my opinion requires a thorough discussion. For example, Funke et al. (2005), their Figure 1, showed much larger NO<sub>2</sub> enhancements than the 10 ppbv shown here.

Reply:

The validation of MIPAS-ESA observations (v 4.61) has been discussed in many papers (Ridolfi et al., 2007; Cortesi et al., 2007, Dethof et al., 2004, see also Ménard et al. 2007, chapter 4, for a review). Moreover, comparison of MIPAS-ESA with MIPAS-IMK has been done very thoroughly and extensively by Wetzel et al. (2007) for NO<sub>2</sub>, which suggested a general good agreement between MIPAS-ESA and MIPAS-IMK (see figure 15, bottom panel, of Wetzel et al paper). However, above 50 km, MIPAS-ESA starts degrading and underestimates values of NO<sub>2</sub> compared to MIPAS-IMK mostly due to non-LTE effects which are included in the latter retrievals. Finally, during SPE, below 30 km, the MIPAS-ESA retrieval could suffer from very large errors (Wetzel et al., 2007). Firstly, in our paper we thus have been careful in not drawing any conclusion for MIPAS operational NO<sub>2</sub> or other constituents above 0.5 hPa (about 50 km), the presence of the model top sponge layer corrupting results at model top in any cases. Secondly, concerning the region below 30 km, no impact were found on our calculation or diagnosis (see figure 7). In any cases, our domain of interest was roughly between 30-50 km of altitude for NO<sub>2</sub>. Wetzel et al. (2007) also presented comparisons with

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ACE, HALOE, SAGE II and POAM II and showed fair to very good results with MIPAS-ESA NO<sub>2</sub>. For temperature, O<sub>3</sub> and HNO<sub>3</sub>, section 2.3 of our paper discusses the validity of MIPAS-ESA (operational) data against different data sets. Discrepancy between the two sets (ESA and IMK) exists but this does not affect our conclusions since NO<sub>2</sub> does not enter into O<sub>3</sub> loss calculation in the context of our methodology (i.e.: passive OmF of ozone only are used for polar ozone loss calculation). Concerning ozone, MIPAS-ESA has been shown to verify well (bias within  $\pm 15\%$ ) in two periods of different EPP strength and in different areas, not in only in the polar regions (Ménard et al., 2007, chapter 10). On the other hand, the assimilation has shown to correctly rectify the model 6-hour predictions for ozone (Figs. 4c and 9c of our paper). Therefore, even if MIPAS-IMK data generally show less random error for many constituents (as shown by our own in-house verification), the fact that MIPAS operational data has better spatio-temporal availability and coverage oriented our choice towards the latter. If our interest, for example, would have been to calculate the mass fluxes or NO<sub>2</sub> budget due to EPP, the use of IMK would have been required. For our purposes here, as long as constituents are reasonably good in the region 30-50 km, they could be used to illustrate our points. We have revised section 2.3 of our original paper following the above discussion and have included a mention about the results obtained by Wetzel et al. (2007) in order to address referee 1's comment. Moreover, to be more specific, we suggest a slight change in the title of the paper to reflect that we have used operational MIPAS data (as opposed to MIPAS-IMK), e.g.: "Impact of energetic particle precipitation on stratospheric polar constituents: an assessment using monitoring and assimilation of operational MIPAS data". There is some uncertainty as to the level of significance of the second part of the above comment: "Funke et al. (2005), their Figure 1, showed much larger NO<sub>2</sub> enhancements than the 10 ppbv shown here". Fig. 1 of Funke et al. (2005) seems to have used MIPAS-ESA data rather than MIPAS-IMK. Also, in our study, OmF are shown and not observations alone as in Funke et al. paper. Finally, the part of Fig. 1 which is pertinent to our paper is that covering August through October where values are in the range 10-20 ppbv. We have added up a sentence in

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the revised version to clarify the spatio-temporal domain of comparison that we have used (see revised section 5.2).

Referee 1:

4. The HNO<sub>3</sub> increases have been attributed to ion-ion recombination, see Verronen et al. (Geophys. Res. Lett. , 2008), unfortunately this paper is not cited. I suggest to review the paragraphs discussing the HNO<sub>3</sub> enhancements in the light of the Verronen paper.

Reply:

We appreciate the comment that the Verronen et al. paper should have been referenced as pointed out. The literature on the subject of HNO<sub>3</sub> enhancements related to EPP periods reveals different possible mechanisms. First, Aikin (1994;1997) and Verronen et al. (2008) claimed that ion-ion recombination is the most important process in the upper stratosphere which explains the HNO<sub>3</sub> enhancement following SPE. However, this process becomes less important in the middle or lower stratosphere at the expense of the ion-cluster chemistry and/or heterogeneous chemistry. On the other hand, Kawa et al. (1995), McDonald et al. (2000), de Zafra and Smyshlaev (2001), Orsolini et al. (2005) and Stiller et al. (2005) also analyzed this enhancement and conclude that ion-cluster chemistry and/or heterogeneous chemistry on sulfate aerosols seem to be the two dominant mechanisms of production of enhanced HNO<sub>3</sub> layers in the stratosphere. In Fig. 8a, we have identified 4 regions of maximum HNO<sub>3</sub> OmF (labeled 1, 2, 3 and 4 on the figure of the original paper). With assimilation (Fig. 8c), the maxima of regions 3 and 4 disappear and only 1 and 2 still remain although somehow reduced. This would indicate that the former are associated with slow time scale errors whereas the latter are linked with faster time scale errors. Since ion-ion recombination is rather a fast time scale process (as compared to water ion cluster chemistry, see Verronen et al.(2008)), it is unlikely that this process be associated with region 3 and 4. On the other hand, in region 1, the presence of ion-ion recombination is important as

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already mentioned in our paper. Finally, in region 2, we agree that ion-ion recombination could also be a likely process. We then have added, in section 5.3 of the revised version, a mention of this possibility and a reference to Verronen et al. (2008) as well as some of the other papers mentioned above.

Referee 1:

Further comments: 1. 22460 I1 Since in this work you present EPP direct and indirect effects, the wording “geomagnetic events occurred which produced massive amount of energetic particles” is misleading. In the direct EPP case, namely solar proton events, the particles mainly originate from the sun/solar wind and are not produced by geomagnetic activity.

Reply:

We agree and have replaced the word “geomagnetic” by simply “EPP” which is more general and includes both direct (SPEs) and indirect (electron precipitation) effects thus eliminating the possible confusion described by referee 1.

Referee 1:

2. 22461 L28 why “in this case”? According to Randall et al., 2006, EPP-IE is always a result of ionized particles trapped in the magnetosphere as correctly described here.

Reply:

The “in this case” has been removed and the sentence re-worded as suggested by another referee (see reply to referee 2).

Referee 1:

3. 22462 L5: Fischer et al, 2007 should probably be Fischer et al, 2008 Reply:

This typographic error has now been corrected.

Referee 1:

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4. 22467 I17: Maybe provide a reference for the sudden warming statement

Reply:

Manney et al. 2005 has been added as reference for the sudden warming period of 2003 in the Northern Hemisphere.

Referee 1:

5. 22474 I7: Why do you not show results for the entire SH winter? The effects of EPP are visible starting in May, maximizing in June and July.

Reply:

We agree that the EPP effects are visible starting in May (see Funke et al., 2005) but mostly above potential temperature of 2000oK (in the mesosphere which is near our model top in the sponge layer). Our interest was in a specific portion of the spatio-temporal domain of 2003 as explained below. There is no necessity to start as soon as May or June 2003 in our context since the focus is limited to the impact of EPP effects on the partial or total ozone column in the stratosphere. Such impact on the ozone column is likely to be negligible from May to July but becomes significant after the polar night is over in the period 280-310 (Sept-Oct) where NO<sub>x</sub> are transported downward to the stratosphere under sunlit conditions. Then NO<sub>2</sub> is converted to NO and the NO<sub>x</sub> catalytic reaction can operate during the period chosen. As well, the HNO<sub>3</sub> anomaly affects the stratosphere but only after the start of our simulations (mid-August 2003). Our model top is 0.1 hPa and the model sponge corrupts the result certainly above 0.5 hPa in polar regions. The tail of the NO<sub>x</sub> intrusion and remnants of the HNO<sub>3</sub> anomalous layer is fully in our model domain far away from the model sponge only in Aug-Oct 2003 and it is also where we can analyze it properly with MIPAS-ESA data.

Referee 1:

6. 22474 I15: How was the end-of-polar night date chosen? Reply:

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The end of the polar night in an absolute way is of course at the equinox. But obviously photochemical processes start before that date in the polar circle and many of the model grid points have already seen the sun. The choice of definition for a given study is also dependent on the nature of the goals achieved. We have selected September 12th (day 255) since beyond this date most of the area beyond 60°S is partially illuminated over the 24 hour period. This date helps to illustrate the period at which the photochemistry starts to have a significant impact on the mean O-P value in the stratospheric region. Following referee 1's query, corresponding text has been added in the revised version specifying that this choice has been drawn from the various panels in order to for facilitate the discussion.

Referee 1:

7. 22482 I10: Please compare this to other work done on this SPE. Vogel et al., 2008, e.g. they found a loss of up to 5.5 DU until the end of November.

Reply:

This is now addressed in the revised version of the paper (section 5.4). First, note that the 5.5 DU result in Vogel et al. (2008) corresponds to a “max NO<sub>x</sub> run” and not a likely scenario. At the end of section 4, Vogel et al. 2008 write: “the comparison between simulated and observed ozone mixing ratios confirms that the max NO<sub>x</sub> run is an upper limit case which overestimates the strength of the NO<sub>x</sub> intrusions. A more realistic value proposed in their paper is 3.3 DU for a daily average value for latitudes greater than 70° (eq. lat.). On the other hand, on our side, the result obtained of 0.91 DU (see table 2 of our paper) corresponds to a 4 weeks average (as compared to a daily average in Vogel et al. paper) and for latitudes greater than 60°N (as compared to 70°N eq. lat. in the Vogel et al. paper). Therefore, our averaging spatio-temporal domain is larger and this represents a different basis of comparison. So in order to compare more adequately with the Vogel et al. paper, we recomputed the polar loss but now for latitudes greater than 70° (instead of 60°) and have taken daily average (in-

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stead of 4 weeks average). The recomputed result obtained for comparison purposes by changing the averaging period and spatial domain is now evaluated to 1.45 DU. This result is fairly close to 1.5 DU obtained by Jackman et al. 2005 (as mentioned by Vogel et al., 2008 in section 5) for the same event. As aside, the Vogel et al. paper refer to “accumulated ozone column” and it is not known if this might imply a difference in definition which might have also contributed to differences in quantities. In any case, as mentioned by Vogel et al. 2008 (end of their section 5) concerning the SPE event of 2003: “on an absolute scale, the differences are small and our results support the conclusion of Jackman et al. (2005) that the impact of solar proton events on the northern hemisphere total polar ozone decreases is small”. We also agree with this statement. Note that another aspect is that the meteorological conditions and the transport which is likely to be different between our system and the system presented by Vogel et al. 2008 might also be a source of differences in results.

Referee 1:

8. 22482 I23: What do you mean with “were found to be unrealistic”? In principle, the OmF technique seems very well suited to do this kind of analysis of the total ozone loss since model dynamics do not change.

Reply:

We agree that the word “unrealistic” may be not clear. That statement has been replaced by: “EPP-IE impacts on ozone are negligible below 30 hPa so justifying the use of partial column (7-30 hPa). Note that the use of total column is not appropriate here since.” (see the original paper for the list of the 3 arguments supporting this sentence). We wish to express that the use of total columns could be misleading since OmF contributions resulting from other factors associated to the lower stratosphere (between 100-30 hPa) tend to have a much bigger weight on the total column as the air density is larger in the lower stratosphere than that in the upper stratosphere. Therefore, OmF not related to EPP in the lower stratosphere could completely mask the impact of EPP.

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This is the reason why we adopted the method of partial columns and not the method of total column differences which could give misleading results in this context.

Referee 1:

9. Unfortunately, the figures provided in the manuscript are not suited for final publication. I suggest to improve the quality (vector graphics) as well as the readability: For the “STD\_xx” plots I suggest to use a different color palette, having the dotted line (referring to 0 in the BIAS\_xx plot) in the middle is misleading.

Reply:

The figures 2,3,4,5,7,8 and 9 have been redone now using graphics software based on IDL. The improvements address the comments of both referees 1 and 2. The color palette has been changed and plots have been saved (and produced) as vector graphics as suggested. Figures have been redone according to the specification of both referees.

Referee 1:

10. The authors use “Julian day” to refer to the day of year. While this use of the term Julian day” is frequent, I suggest to replace it by the correct term “ordinal date” or “day of year”, or, even better, use day/month labeling

Reply:

This has been changed to “day of the year”. We found that this labeling makes it easier and provides a more accurate reference of the different events described in our paper.

Referee 1:

11. 22474 l22: Typo “persist persisting”.

Reply:

This has been corrected.

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We added the following list of new references in the revised version. For other references mentioned in our reply above please refer to the ACPD submitted paper. Note that an additional reference dealing with EPP has also been added for completeness (e.g. Baumgaertner et al., 2009).

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Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/9/C9859/2010/acpd-9-C9859-2010-supplement.pdf>

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