Referee #1:

Article summary: The authors present an analysis of energetic particle precipitation effects on the polar middle atmosphere. The impact of low (auroral) and middle (radiation belt) energy range electrons on polar reactive nitrogen (NOy), reactive hydrogen (HOx), and ozone are quantified. They used the chemistry climate model SOCOL3-MPIOM and Envisat Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) and Aura Microwave Limb Sounder (MLS) satellite instruments in their studies. By comparing a year of high electron precipitation with a several year-long quiescent period, they found that there was about 15% less ozone in the polar stratosphere during Southern Hemisphere winter in both model predictions and satellite observations.

Review summary: The paper has some very interesting results on the impact of energetic particle precipitation on the polar middle atmosphere. In particular, they quantify the effect of low and middle energy range electrons with both a model and satellite instrument measurements. This type of study of the atmospheric impact of electrons in different energy ranges has only been considered before in a few other studies. The paper provides interesting results for the rather active year 2005 in comparison with the more quiescent time period 2006-2010. I do think that the paper should be published. The paper is generally well-written, but I have some suggested wording changes.

Suggested wording changes:

1) Page 2, line 13: Change “found a significant” to “found significant”

2) Page 3, line 13: Change “nitrogen monoxide” to “nitrogen monoxide (nitric oxide)”

3) Page 3, line 31: Change “aboard Aura” to “aboard the Aura”

4) Page 5, line 6: Change “brings model” to “brings the model”

5) Page 5, line 7: Change “from outer” to “from the outer”

6) Page 5, line 10: Change “In study” to “In the study”

7) Page 5, line 13: Change “analyzed 2005” to “analyzed the 2005”

8) Page 5, line 18: Change “in JJA” to “in the JJA”

9) Page 6, line 2: Change “simulation mesospheric” to “simulation the mesospheric”

10) Page 6, line 11: Change “Recent study” to “A recent study”

11) Page 6, lines 20-21: Change “in LEE, MEE and REF simulation” to “in the LEE, MEE and REF simulations”

12) Page 7, line 6: Change “Second difference” to “A second difference”

13) Page 7, line 7: Change “for small” to “for a small”

14) Page 7, line 9: Change “by increase” to “by an increase”

15) Page 7, line 9: Change “small increase” to “small increases”

16) Page 7, line 15: Change “shows increase” to “shows an increase”

17) Page 7, line 15: Change “cause increase” to “cause increases”

18) Page 7, line 16: Change “or tenth” to “or a tenth”

19) Page 7, line 17: Change “total” to “the total”

20) Page 7, line 21: Change “ALL case” to “the ALL case”

21) Page 7, line 27: Change “albeit” to “albeit the”

22) Page 7, line 27: Change “Biggest” to “The biggest”

23) Page 7, line 30: Change “300” to “300 ppbv”

24) Page 7, line 31: Change “Since sum” to “Since the sum”

25) Page 8, line 1: Change “for ozone” to “for the ozone”

26) Page 8, line 3: Change “is more” to “is the more”

27) Page 8, line 3: Change “produce stratospheric” to “produce the stratospheric”

28) Page 8, line 13: Change “to reproduce” to “to the reproduction of”

29) Page 8, line 24: Change “near HOx” to “near the HOx”

30) Page 8, line 25: Change “and increase HOx” to “and an increase of HOx”

We would like to sincerely thank the reviewer for their suggestions. All their points have been included in the text.

Referee #2:

We thank the reviewer for their constructive suggestions that contributed to improving the quality of this paper. We have carefully analyzed and addressed all comments below.

A straightforward paper which is timely for the discussion of the topic in the community.

I have only few concerns being major, and otherwise recommend the paper for publication if my concerns have been clarified.

1. The authors use a flux boundary condition for including the NOy produced in the upper mesosphere and lower thermosphere. They criticize the alternative method of prescribing mixing ratios as possibly inconsistent. I do not agree with this general statement. In my opinion, this depends on the dynamical boundary conditions of the model. At the ground you may introduce an influx of some species essentially via a turbulent flux, but in the mesosphere the influx of NOy is by an advective term. Setting the influx from the parameterization, you may have an increase in concentration even when having upwelling which is physically impossible.

We have rephrased our statement to emphasize its relevance to our particular model:
"Prescribing concentrations requires overwriting NO simulated values. It is inconsistent with the treatment of the physical and chemical processes in our model leading to accumulation of NO_y. This is not the case for influx approach and therefore we prescribe the NO influx instead of mixing ratios, however prescribed NO mixing ratio can be used for models with different treatment of the chemical and transport processes."

2. The chosen periods (2005 for high and 2006 - 2010 for low activity) should be explained. 2003 (at least 50% higher Ap) and 2008-2009 could have been a better choice. As the Halloween storm occurs in late 2003, this event should not interfere for your study. In addition MIPAS' coverage in 2005 is not as good as in other years. As MLS data are not available for 2003, MIPAS ozone data could be used.

We chose 2005 for high and 2006 - 2010 for low geomagnetic activity because we want to include two satellite datasets for our study and ozone measurements of MLS instrument start at year 2005. Moreover, it is true that MIPAS' coverage is not as good as in the other years, but the lack of observations appears only in September. This paper focuses on JJA period which is well covered with MIPAS in year 2005 and choosing this period allows us to use both satellite instruments (MIPAS and MLS) in the same manner. To clarify we added in the first paragraph of section 3.1:

"Even though year 2003 on average has higher Ap, here we choose year 2005 as the geomagnetically active year. This allows us to compare modeled NOy and ozone using two different satellite datasets MIPAS and MLS (which is available only since 2005). MIPAS data are unavailable from September 2005 to the end of the year, but our main period of interest is JJA, which is well covered by the observations."

3. The pronounced mesospheric minimum of the NOy concentration in the REF runs during SH mid-winter needs an explanation. The mentioned SPEs should show up in the middle mesosphere in the whole SH winter. Perhaps you could provide a figure with a higher time resolution as you have done for ozone.

SP events should be visible as NOy enhancements, but unfortunately due to the limited storage space, we don't have a higher temporal resolution for NOy as we do for ozone.

4. Why does the SP event of June 2005 does not show up in Fig 3?

SP of June 2005 was one of the minor events (M4; ftp://ftp.swpc.noaa.gov/pub/indices/SPE.txt). Although even smaller event is seen on Figure 3 (M3, end of July), it is possible that the June event was lost due to the dynamic variability.

5. Please provide an additional figure with an mesospheric transport tracer as for example CO (active - ref years), for comparison to exclude or evaluate dynamical effects.

Please see the attached plots. Although there is up to 10% less CO in the year 2005 compared to the 2006-2010, the difference is not statistically significant. Our study focuses on chemical effects of EPP- Follow-up study will focus with temperature and dynamics caused by EPP.

We added in the following sentence in the Conclusions, second paragraph: "Future work is required to address the roles of indirect changes in temperature and dynamics in the EPP-induced stratospheric ozone variation."

6. Yet technically, but nevertheless important for the understanding of possible effects in the lower stratosphere, the colors in Fig 2 do not really allow to decide where small values are significant. Please use a different color table.

We changed the colormap of the Figure 2 and reduced the number of plotting levels to make the plot clearer. We did the same change for NOy in Figure 4 to keep the plotting style consistent.

7. The enhancement of NOy by MEE (Fig4) outside the polar vortex needs some discussion of the photochemical lifetime expected outside the polar vortex, in sunlight.

We added the following sentence in the second paragraph in the section 3.3:

“Around 0.01 hPa, EPP produced NOy increases from 50 ppbv at around 60° S, where NOy lifetime is decreased due to the sunlight, to more than 500 ppbv at the pole, in the polar night.”

8. A main result of the paper is the impact of MEE, essentially via HOx, on ozone which the authors estimate to be of the same order as NOy produced by LEE. This is important for the understanding of EPP effects, but this result needs in my opinion more substantiation. The fact that MEE in 2005 mostly come with SPEs (Fig. 3) reminds me that there were some discussions about crosstalks of the detectors for the different particles especially in the MEE energy range. For example, Anderson et al. 2012, exclude electron fluxes during SPEs in their analysis because of possible contamination. Please try to extend your analysis when excluding SPE periods.

We addressed the issue about proton contamination of electron channels during SP events. In section 3.1 in 4th paragraph we added:

“During strong SP events protons can contaminate the highest electron channel, so this channel is excluded from the AIMOS dataset (Yando et al 2011). Although some degree of contamination is still possible in the lower channels, protons are not the sole cause of the increased NOy in this SP event. Namely, SP events are often associated to large coronal mass ejections that form a shock in front of them. Once the shock hits the Earth if often leads to a geomagnetic storm which leads to acceleration of electrons of > 30 keV energies. Therefore, increased MEE precipitation often happens very shortly after SP event because the shock and the geomagnetic storm are related to the same coronal mass ejection driver (Asikainen and Ruopsa 2016).”

By excluding SP events we would not get valid comparison with satellite measurements (Fig 2 and Fig 3) in which all EPP are present. Because Fig 4 shows difference of experiment (ALL, LEE and MEE), where we have SP, and REF run, which also contains SP, we can focus on electron influence only.

9. The authors should also improve the grammar of their paper with a special emphasis on the use of articles. Minor comments are marked in the commented pdf attached. Please also note the supplement to this comment: https://www.atmos-chem-phys-discuss.net/acp-2018-1123/acp-2018-1123-RC2- supplement.pdf

Thank you for the corrections, please see the answers below.

1. “or chemistry transport models”

Corrected.

2. “auroral only?”

Yes, Baumbaertner et al (2009) parameterization deals with low energy electrons (auroral) which are prescribed as NO influx through the model top.

3. “reference uses just one model”

Corrected. We removed “several”

4. “give reference”

Corrected. We added Rozanov et al 2012

5. “no new paragraph. Please combine”

We removed the last sentence of the first paragraph so separation is now better. The first paragraph describes new LEE parameterization, while the second presents the aim of the paper.

6. “ionization by … is”

Corrected.

7. “I do not see how the approaches really differ. This may depend on the implementation: could you please specify?”

The first approach is prescribing NO in the model domain between 0.09 – 0.01 hPa and the second is prescribing influx through the model top. We added “instead of fluxes through the model top” to clarify.

8. “Why this periods? 2005 is not the best year for MIPAS coverage. I propose to check 2003 (esp. first half) and 2008 - 2010.”

We have already addressed this comment above.

9. “in the MIPAS observations”

Corrected.

10. “Somewhat frustrating for the reader that the authors do not try to answer by themselves.”

This is true, but this issue deserves a study on its own, which is beyond the scope of this paper.

11. “style”

This is not clear, but we’ve changed the sentence to: ” The modeled NOy mesospheric anomaly peak is absent and enhancement of 10 ppbv…”

12. “alone”, “the”, “was”

Corrected.

13. but you are showing diffs between high and low years too.??

We removed “and not by on/off experiments as done here”

14. “specify”

The difference is very small and not statistically significant. We changed “small” to “does not show a statistically significant”

15. “compared”

Corrected.

16. “Please explain missin June-16 event”

We have addressed this comment above. This event, because of its small magnitude was probably lost in the dynamic variability. However, the absence or presence of the SP event in question will not change the conclusions of the study.

17. “mean”

Corrected.

18. “combine sentences”

Combined.

19. “Do you really mean equatorward”

We changed this to “Between 30-35° S… “

20. “the”

Corrected.

21. “unclear. Please be concise”

We changed this sentence to:

“However, increased MEE precipitation coincident with SP events may be a significant contribution to the observed NOy amounts.”

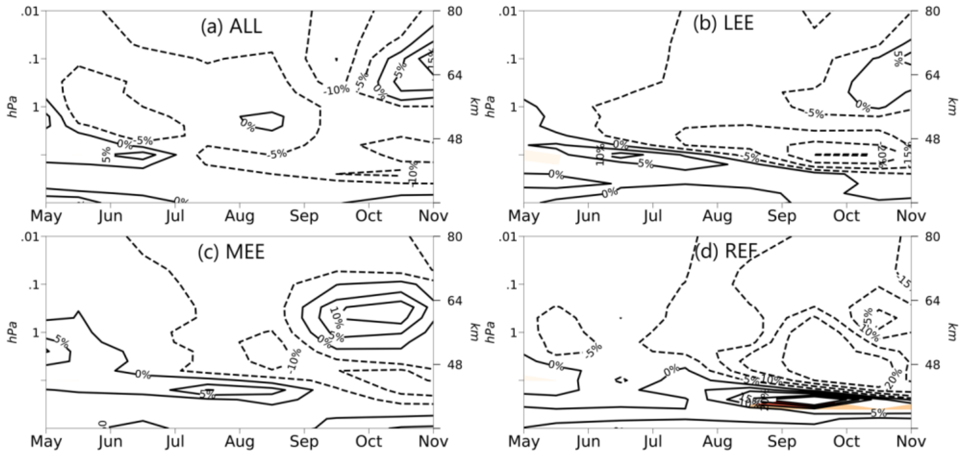

Figure S1: Monthly mean CO volume mixing ratio in % for the Southern Hemisphere (> 70° S average) calculated as difference of the year 2005 and the average 2006 – 2010. (a) ensemble mean of ALL simulations; (b) ensemble mean of LEE simulations; (c) ensemble mean of MEE simulations; (d) ensemble mean of REF simulations. Contour levels are: -20, -15, -10, -5, 0, 5, 10, 15 and 20 %. Colored regions are significant at the 99 % confidence level (calculated using a Student t-test).

Referee #3:

We are sincerely grateful to the reviewer for their contribution. Many of their comments significantly improved our study. We have carefully analyzed and addressed all their comments below.

In the paper “Reactive nitrogen (NOy) and ozone responses to energetic electron precipitation during Southern hemisphere winter” by Arsenovic et al, model studies for the years 2002-2010 incorporating the impact of different populations of precipitating particles (medium-energy electrons, low energy electrons, solar proton events, and all) are analysed. Medium-energy electrons (MEE) and solar protons (SP) are included by prescribing ionization rates, low-energy electrons (LEE, representing the auroral input) are input as an influx of NOy at the model top (1 Pa). Results show generally best agreement with observed NOy and ozone loss when all precipitating particles are included. Medium-energy electrons only contribute a small amount to the stratospheric and mesospheric NOy budget, which is dominated by low energy electrons; however to reproduce the stratospheric ozone loss, both low energy and medium energy electrons are necessary, though the reason for this is not clear. Energetic particle precipitation is considered part of the solar forcing of the climate system, and the LEE parameterization used here is recommended for the CMIP6 model experiments (see Matthes et al 2017). A comparison of model results using this parameterization with observations of NOy and ozone as presented here is therefore of great interest, e.g., for the interpretation of the CMIP6 runs, as is the determination of the individual contributions of the different particle sources as provided here by analysing model runs incorporating MEE, LEE and SP combined and individual. However, I have two concerns which must be considered before publishing. Both will need some changes to the text but no further analysis, so are probably minor, though important in my opinion:

1. In my opinion, the comparison of the individual contributions of MEE versus LEE is the most important result of the paper, as the LEE impact only has already been studied in earlier model studies (e.g., in Sinnhuber et al 2018, see also next comment). Maybe you could emphasize this point more by including this in the title of your paper? However, stating that you compare MEE and LEE contributions is misleading, because the upper boundary of your model – 1 Pa, about 80 km – does not represent a sharp boundary between LEE and MEE. Rather, the MEE impact as defined by you (electrons with 30-300 keV) extends from 100 km to possibly below 70 km altitude, while the LEE impact (aurora) probably extends from around 90 km upwards. There are three obvious difficulties with separating the individual impacts of LEE and MEE here: a) there is some overlap of MEE and LEE between 90-100 km; b) MIPAS observations, on which the upper boundary conditions for LEE are based, only scan the atmosphere up to 68 km altitude, so implicitly include contributions from both MEE and LEE which cannot be separated; c) the upper boundary of your model does not include the whole altitude range impacted by MEE (from 100 km down to probably below 70 km), rather it cuts around 80 km (1 Pa), which might be even below the maximum of the MEE impact (I would assume that this is in the range 80-95 km altitude, though this is not really clear yet). So what you actually investigate are the individual impacts of electron precipitation to above and below your model top, which is interesting in

itself, but not quite the same as the individual contributions of MEE and LEE with the definition provided in your paper. I don’t see how you could do the analysis better, and considering that 1 Pa is rather conventional as upper boundary for state-of-the art CCMs, this is a very interesting investigation anyway. However, you only provide a lower limit for the MEE impact, and you must discuss this in some detail.

We have added two sentences in second paragraph, describing altitudes of auroral and radiation belt electrons: Radiation belt electrons (energies > 30 keV) impact chemistry below 90 km in the atmosphere (Turunen et al 2009)…. [LEE] Their peak impact is above 90 km in the thermosphere (Turunen et al 2009).

Methods, 3rd paragraph added sentence: “As mentioned before, LEE precipitate above 90 km and MEE precipitate between 70 and 90 km altitude (Turunen et al 2009). However, because of our model top at 80 km, here we consider electrons that precipitate below 80 km as MEE and electrons that precipitate above model top as LEE.”

2. There is one publication (Sinnhuber et al 2018) discussion results of model experiments using the same upper boundary condition for LEE NOy as used here, considering the same period of time (2002-2010), and using a model with the same top height (1 Pa = 0.01 hPa) with probably very similar model dynamics (both based on ECHAM5). Results of this model experiment are compared to NOy and observed ozone loss, and generally show a good agreement. So you really must emphasize what is new in your paper compared to Sinnhuber et al 2018. In my opinion this is: a) you use a different setting of the upper boundary condition, incorporating fluxes instead of VMR, and b) you investigate the individual contributions of NOy above and below your model upper boundary. Concerning the first point, you should compare your model NOy and ozone loss to the results of Sinnhuber et al 2018, to investigate whether there is an obvious difference between including NOy flux or VMR. My impression being that results are probably very similar for NOy, but mesospheric ozone loss seems more realistic when using flux over the model top boundary compared to VMR.

We added sentence in the last paragraph in the introduction:

Sinnhuber et al (2018) showed impact of this LEE parameterization in their EMAC model on NOy and ozone, they used another method of prescribing LEE and they don’t consider MEE.

We added Sinnhuber et al (2018) citation in methods “Matthes et al (2017) and Sinnhuber et al (2018) also implemented…”

Comparison of Sinnhuber et al. (2018) is given in results, section 3.1:

“Sinnhuber et al (2018) showed similar results in EMAC and KASIMA models and overestimation of NOy in 3dCTM model in southern hemisphere compared to MIPAS observations.”

In the last paragraph of section 3.2 we added: “In the study of Sinnhuber et al (2018) the three analyzed models (3dCTM, KASIMA and EMAC) generally show good agreement with the satellite observations.”

3. Page 2, lines 9-10: the recommendation is either as influx through the model top or as volume mixing ration in the upper model boxes. Please clarify.

We have changed this sentence as suggested: “For climate models that have an upper lid below the thermosphere, a prescription of LEE either as NOx influx through the model top or as volume mixing ration in the upper model boxes is recommended (Matthes et al 2017).”

4. Page 2, lines 16 to 23, description of the MIPAS-bases upper boundary condition: my understanding is that MIPAS scans the atmosphere up to 68 km altitude, so any parameterization based on MIPAS data has to be extrapolated to the upper mesosphere above ~70 km. Evidence (from previous usage of this upper boundary condition) seems to be that this works really quite well, but nevertheless it must be a restriction of the method if applied to around 80 km altitude (1 Pa) as you do, and you must discuss this. Also, because the upper boundary condition is based on observations of the mid-to lower mesosphere, it is not a parameterization of LEE only as stated here – it must be a mixture of LEE and MEE effects, as these are not clearly separated in the observations in any way – it is NOy reacting correlated to changes in the geomagnetic indices, possibly with some time-lag, in both cases.

Regarding the MIPAS scanning the atmosphere to 68 km, we added the following text:

Methods, 3rd paragraph: “Although MIPAS scans the atmosphere up to 68 km altitude, the applicability of this parameterization above 70 km has been validated by comparing to MIPAS Middle and Upper atmosphere observations (scanning up to 100 and 170 km, respectively).”

It is true that MIPAS shows the mixture of LEE and MEE NOy, but it is impossible to clearly distinguish between them and this approximation is inevitable. However, NOy coming from MEE has a very small contribution to total NOy compared to LEE (as we showed in this paper), so this approximation should not have a big impact on ozone loss and thus induced changes.

5. Page 2, lines 22-23: it has already been shown in Sinnhuber et al 2018 that this upper boundary condition works really well in reproducing mesospheric and stratospheric NOy. This was actually done for exactly the same period of time as investigated in your paper (2002-2010), and using a model also based on ECHAM5 (EMAC), so you shouldn’t generalize so much here (this already has been done), instead focus on what really is new in your paper: a) showing that the parameterization also works well if used as a flux through the upper boundary, and b) investigating the individual contributions of NOy above and below your model upper boundary. Please clarify.

We have removed line “It is therefore important to demonstrate that the particle impact is well represented in chemistry-climate models.” from the introduction and added:

“Although Sinnhuber *et al* (2018) showed impact of the new Funke *et al* (2016) parameterization in their EMAC model on NO_y and ozone, they used another method of prescribing LEE and they did not consider the MEE. Here we present results from our state of the art chemistry-climate model that employs the same parameterization of LEE together with the previous representations of other energetic particles.” to the last paragraph in the introduction.

6. Page 3, line 9: A note on the definition of MEE and LEE here. As pointed out already above, the MIPAS upper boundary will probably be affected by MEE as well, as it is based on observations below 70 km altitude. On the other hand, AIMOS data in the vertical range of your model probably do not include all MEE effects even in the definition used here (30-300 keV electrons), as 30 keV electrons will have an impact on the atmosphere above your model boundary (1 Pa ∼ 80 km?) as well. Actually I would expect MEE to have the largest impact on NO in the altitude region 80-95 km. Actually I think that your model experiments with and without EEP NOy above and below your model lid are very interesting; however, I also think that you should clarify that this is not exactly the same as separating auroral and MEE electrons, due to the altitude of your model lid, and because there must be a vertical area where both overlap (probably between 80-100 km), and contributions are separated neither in AIMOS, nor in the MIPAS-based upper boundary condition. So please clarify this here.

We added to Methods section in 3rd paragraph: “As mentioned before, LEE precipitate above 90 km and MEE precipitate between 70 and 90 km altitude (Turunen et al 2009). However, because of our model top at 80 km, here we consider electrons that precipitate below 80 km as MEE and electrons that precipitate above model top as LEE.”

7. Page 3, line 30-31: please state the vertical resolution (not the same as the vertical spacing of the limb-scans) of MIPAS NOy, and make some statement about the applicability of NOy above the top altitude of the MIPAS nominal limb scans.

The vertical resolution of MIPAS NOy from the cloud top altitude up to 70 km is 3-5 km.

We have edited the sentence to: ”Since it provides the entire NOy budget in the upper atmosphere (with vertical resolution of 3-5 km), we used this dataset to validate simulated NOy.”

Regarding the second part of the comment, in the last paragraph of Methods section, we added: “The top altitude of the MIPAS nominal limb scans is 68 km, but it also contain information on the NOy above, though with low vertical resolution.”

And in Methods section in the 3rd paragraph:

"Although MIPAS scans the atmosphere up to 68 km altitude, the applicability of this parameterization above 70 km has been validated by comparing to MIPAS Middle and Upper atmosphere observations (scanning up to 100 and 170 km, respectively)."

8. Page 4, lines 12-26, discussion of Figure 2: the comparison of the different model results to the observations is actually quite difficult, because a) the observations do not cover the whole year, b) NOy vmr is plotted with a logarithmic scale using shades of red – it really is quite easy to overlook a factor 2 difference in that way. I guess you had a good reason to choose 2005 as sole "high Ap" year. However, you could improve on b), maybe by using a color-scale with more contrast, probably also by overplotting the 1, 10, 100 and 1000 ppb contours of MIPAS on the different model scenarios. That would really help to distinguish differences in timing and magnitude, at least qualitatively, in a more comprehensive way.

We address the lack of data by adding the following sentences in the first paragraph in the section 3.1:

"Even though year 2003 on average has higher Ap, here we choose year 2005 as the geomagnetically active year. This allows us to compare modeled NOy and ozone using two different satellite datasets MIPAS and MLS (which is available only since 2005). MIPAS data are unavailable from September 2005 to the end of the year, but our main period of interest is JJA, which is well covered by the observations."

Regarding the Figure 2, we changed the colormap and reduced the number of plotting levels to make the Figure clearer.

9. Page 4, line 19 and line 20: the model and upper boundary condition discussed in Matthes et al 2017 and Sinnhuber et al 2018 are actually the same (though possibly a newer version of the upper boundary condition), so this sounds like a contradiction – NOy is underestimated in Matthes et al 2017 and overestimated in Sinnhuber et al 2018?

We have corrected the sentence in question:

"Sinnhuber et al (2018) showed similar results in EMAC and KASIMA models and overestimation of NOy in 3dCTM model in southern hemisphere compared to MIPAS observations."

10. Page 4, line 25-26, "This difference is coming from increased MEE precipitation . . ." but is this real (compared to observations) or does it rather suggest a problem of the ionization rates used? Possibly due to proton contamination of the electron detectors during strong SP events?

We have added to the 4th paragraph in the section 3.1: "During strong SP events protons can contaminate the highest electron channel, so this channel is excluded from the AIMOS dataset (Yando et al 2011). Although some degree of contamination is still possible in the lower channels, protons are not the sole cause of the increased NOy in this SP event. Namely, SP events are often associated to large coronal mass ejections that form a shock in front of them. Once the shock hits the Earth if often leads to a geomagnetic storm which leads to acceleration of electrons of > 30 keV energies. Therefore, increased MEE precipitation often happens very shortly after SP event because the shock and the geomagnetic storm are related to the same coronal mass ejection driver (Asikainen and Ruopsa 2016)."

11. Page 5, lines 7-8: the possible contribution of MEE to mesospheric NOy during SPEs has already been investigated (using AIMOS data) in Wissing 2010 (also in Funke et al 2011?). However, if I remember correctly, the rather large impact discussed in Wissing 2010 might in part be due to proton contamination of the electron channels. How sure are you that this is not the case here?

We have addressed this comment in the previous answer.

12. Page 5, lines 22-24: how do your results compare to the model results of Sinnhuber et al 2018?

We added citation to Sinnhuber et al (2018) in this sentence.

13. Page 6, line 31: how does the ozone loss compare to Reddmann et al 2010?

The comparison with Reddmann et al (2010; Figure 14a) is not possible because they show different time period (single year 2003, where the Ap index is very high) and they show vertical propagation in time, while we discuss averaged JJA period.

14. Page 8, line 2: the transport of large amounts of NOy, I assume.

Although NOy produced by LEE gets transported in the downwelling circulation, LEE affect the chemistry by producing the large amounts of NOy.

15. Page 8, line 18-19: though the reason is not quite clear.

We added the sentence: "Future work is required to address the roles of indirect changes in temperature and dynamics in the EPP-induced stratospheric ozone variation."

Reactive nitrogen (NO_y) and ozone responses to energetic electron precipitation during Southern Hemisphere winter

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Abstract. Energetic particle precipitation (EPP) affects the chemistry of the polar middle atmosphere by producing reactive nitrogen (NO_y) and hydrogen (HO_x) species, which then catalytically destroy ozone. Recently, there have been major advances in constraining these particle impacts through a parametrization based on high quality observations. Here we investigate the effects of low (auroral) and middle (radiation belt) energy range electrons, separately and in combination, on reactive nitrogen and hydrogen species as well as on ozone during Southern Hemisphere winters from 2002 to 2010 using the chemistry-climate model SOCOL3-MPIOM. Our results show that, in absence of solar proton events, low energy electrons produce the majority of NO_y in the polar mesosphere and stratosphere. In the polar vortex, NO_y subsides and affects ozone at lower altitudes, down to 10 hPa. Comparing a year with high electron precipitation with a quiescent period, we found large ozone depletion in the mesosphere; as the anomaly propagates downward, 15 % less ozone is found in the stratosphere during winter, which is confirmed by satellite observations. Only with both low and middle energy electrons, our model reproduces the observed stratospheric ozone anomaly.

1 Introduction

Energetic particles originating from the Sun, the magnetosphere, or from outside the solar system continuously precipitate into the Earth's atmosphere and can influence atmospheric processes. They ionize neutral air molecules especially in the middle and upper polar atmosphere and create odd nitrogen and hydrogen species, NO_x ([N] + [NO] + [NO_2]) and HO_x ([H] + [OH] + [HO_2]). NO_x and HO_x radicals can catalytically deplete ozone. The *in-situ* destruction of ozone in the mesosphere is characteristic for HO_x due to its fast reaction rates (Bates and Nicolet, 1950). On the other hand, NO_x, in the absence of sunlight, subsides within the down-welling branch of the overturning circulation, affecting ozone concentrations at lower altitudes (Solomon et al., 1982).

High energy particles, i.e. solar protons (Jackman *et al* 2008) and radiation belt electrons (Arsenovic et al., 2016; Semeniuk et al., 2011) can penetrate directly into the mesosphere and stratosphere. Radiation belt electrons (energies > 30 keV) impact chemistry below 90 km in the atmosphere (Turunen et al., 2009). Electrons of lower energies (< 30 keV, auroral) originate from the magnetosphere as well as the radiation belt electrons (Mironova et al., 2015), but they get accelerated in the magnetotail and precipitate into the lower thermosphere in the auroral ovals (55 – 70° geomagnetic latitude) (Baker et al., 2001; Barth et al., 2003). Their peak impact is above 90 km in the thermosphere (Turunen et al., 2009).

There have been numerous attempts to include low energy electrons (LEE) in climate models. Chemistry-climate or chemistry-transport models with top in the thermosphere, e.g. HAMMONIA (Schmidt et al., 2006), KASIMA (Reddmann et al., 2010) and WACCM (Andersson et al., 2018; Marsh et al., 2007), have included effects of LEE directly because they deposit their energy within the model domain. For climate models that have an upper lid below the thermosphere, a prescription of LEE either as NO_x influx through the model top or as volume mixing ration in the upper model boxes is recommended (Matthes et al., 2017). Baumgaertner *et al* (2009) has developed a parameterization of this flux based on the geomagnetic activity A_p index, a daily worldwide measure of the effects of solar wind on the Earth magnetic field. When incorporated into ~~several~~ chemistry-climate model~~s~~, results showed significant ozone depletion in the mesosphere and stratosphere (Baumgaertner et al., 2011). For the SOCOL chemistry-climate model Rozanov *et al* (2012) also found ~~a~~ significant ozone decreases in the mesosphere and stratosphere, with peak values around 10 % in September around 36 km altitude over the Antarctic.

Funke *et al* (2016) have recently developed a semi-empirical model that calculates concentrations and fluxes of mesospheric and stratospheric NO_y compounds ($[NO] + [NO_2] + 2\times [N_2O_5] + [HNO_3] + [ClONO_2]$) based on the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) observations. The model exploits the nearly linear relationship in the mesosphere between A_p index with observed NO_y produced by EPP. This advance in the representation of LEE in climate models motivates us to investigate if LEE can have a larger impact on atmospheric chemistry than previously thought (Rozanov et al., 2012). Moreover, this LEE parameterization is a part of the recommended solar forcing dataset for climate models within the upcoming Coupled Model Intercomparison Project Phase 6 (CMIP-6, Matthes *et al* 2017). ~~It is therefore important to demonstrate that the particle impact is well represented in chemistry-climate models.~~

It is crucial to have a realistic representation of EPP in models as the introduced signal impacts atmospheric chemistry and potentially regional climate (Baumgaertner et al., 2011; Maliniemi et al., 2014; Rozanov et al., 2012; Seppälä et al., 2013). Although Sinnhuber *et al* (2018) showed impact of the new Funke *et al* (2016)~~this~~ ~~LEE~~ parameterization in their EMAC model on NO_y and ozone, they used another method of prescribing LEE and they ~~didn't~~did not consider the MEE. Here we present results from ~~a~~ our state of the art chemistry-climate model that employs ~~the new Funke *et al* (2016)~~ the same parameterization of LEE together with the previous representations of other energetic particles. ~~We compare our results with the satellite observations.~~ This paper focuses on evaluating NO_x and ozone response to LEE and MEE precipitation, separately and in combination, in Antarctic winters (JJA: June, July and August), in order to avoid the more complicated Arctic polar

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vortex with its high variability and strong dependence on meteorological conditions (Hitchcock et al., 2013). We compare our results with the satellite observations.

2 Methods

We used the coupled chemistry-climate model SOCOL3-MPIOM (Muthers et al., 2014; Stenke et al., 2013). The atmospheric dynamic component of the model is ECHAM5.4 (Roeckner and Bäuml, 2003), coupled to the air chemistry module MEZON (Egorova et al., 2003; Rozanov et al., 1999) and the interactive ocean module MPIOM (Jungclaus et al., 2006; Marsland et al., 2002). We carried out the experiments with T31 spectral resolution on 39 vertical levels from the surface up to 0.01 hPa (~80 km).

The model boundary conditions and parameterizations are identical to those described in Arsenovic *et al* (2016), except for the LEE parameterization. Following Calisto *et al* (2011), galactic cosmic rays (GCR) are parameterized as a function of geomagnetic latitude, pressure and solar modulation potential. Ionization by solar protons (SP) is treated according to Jackman *et al* (2008) and ionization by middle energy electrons (MEE) with energies between 30 and 300 keV ~~are~~ is taken from the Atmospheric Ionization Module Osnabrück (AIMOS) v1.6 (Arsenovic et al., 2016; Wissing and Kallenrode, 2009). Electrons of energies higher than 300 keV are not included in the model due to a lack of adequate parameterization.

For LEE, we are using the semi-empirical model for NO_y influx by Funke *et al* (2016) through the model top at 0.01 hPa (75-80 km in polar conditions). Although MIPAS scans the atmosphere up to 68 km altitude, the applicability of this parameterization above 70 km has been validated by comparing to MIPAS Middle and Upper atmosphere observations (scanning up to 100 and 170 km, respectively). As more than 99 % of the NO_y at this altitude is in the form of nitrogen monoxide (nitric oxide), NO (Brasseur and Solomon, 2005), we approximate the NO_y influx calculated by the semi-empirical model as NO influx at this level in SOCOL3-MPIOM. As mentioned before, LEE precipitate above 90 km and MEE precipitate between 70 and 90 km altitude (Turunen et al., 2009). However, because of our model top at 80 km, here we consider electrons that precipitate below 80 km as MEE and electrons that precipitate above model top as LEE.

Matthes *et al* (2017) and ~~(~~Sinnhuber *et al* (2018) also implemented the parameterization by Funke *et al* (2016) in the EMAC model. They used a different approach, prescribing NO concentrations (instead of fluxes through the model top) in the model within the 0.09 - 0.01 hPa layer and performed the simulations with specified dynamics. Prescribing concentrations requires overwriting NO simulated values. It is inconsistent with the treatment of the physical and chemical processes in our model leading to accumulation of NO_y. This is not the case for influx approach and therefore we prescribe the NO influx instead of mixing ratios, however prescribed NO mixing ratio can be used for models with different treatment of the chemical and transport processes.~~Prescribing concentrations requires overwriting NO values during the model run and might be inconsistent with modeled background atmospheric state and the treatment of the physical and chemical processes in the model. This is not the case for influx approach and therefore we prescribe the NO influx instead of mixing ratios.~~

Figure 1 shows the monthly mean geomagnetic A_p index that covers our simulated period. Period 2002-2005 was characterized by a rather high A_p index and the 2006-2010 period by low values. For our simulations, we have used daily NO fluxes calculated from daily A_p indices.

Four sets of 6-member ensemble simulations were carried out, covering the 2002-2010 period: the “ALL” simulation, that includes all energetic particles (GCR, SP, MEE and LEE), the “LEE” simulation (GCR, SP and LEE), the “MEE” simulation (GCR, SP and MEE) and the reference, “REF” simulation (GCR and SP). All these simulations have the same model boundary conditions and differ only in the inclusion of the low/middle energy electron precipitation.

We used two satellite datasets to evaluate our model results: MIPAS for nitrogen species and the Microwave Limb Sounder (MLS) for ozone. MIPAS was a Fourier transform spectrometer aboard the ENVISAT satellite (Fischer et al., 2008). The quality of MIPAS NO_y and individual NO_y species has been extensively assessed in SPARC (2017), as well as specific validation studies (e.g. Bender *et al* 2015; Sheese *et al* 2016). The top altitude of the MIPAS nominal limb scans is 68 km, but it also contains information on the NO_y above, though with low vertical resolution. Since it provides the entire NO_y budget in the upper atmosphere (with vertical resolution of 3-5 km), we used this dataset to validate simulated NO_y.

-The MLS aboard the Aura satellite (Waters et al., 2006) provided daily measurements of ozone profiles (Froidevaux et al., 2008) in the middle and upper atmosphere since August 2004. We used MLS observations to evaluate modeled ozone. The vertical resolution of MLS O_3 (v4.2) is about 3 km in the stratosphere, increasing up to about 5 km in the mesosphere (Livesey *et al* 2018).

3 Results

3.1 NO_y enhancement propagation

Figure 2 shows the difference in NO_y concentration between the geomagnetically active year 2005 and the mean over the geomagnetically quiescent period 2006 – 2010 averaged over 70 – 90°S. Even though year 2003 on average has higher Ap, here we choose year 2005 as the geomagnetically active year. This allows us to compare modeled NOy and ozone using two different satellite datasets MIPAS and MLS (which is available only since 2005). MIPAS data are unavailable from September 2005 to the end of the year, but our main period of interest is JJA, which is well covered by the observations.

The MIPAS observations (Figure 2a) show a NO_y enhancement throughout the mesosphere and upper stratosphere. In terms of mixing ratio, the highest increase of 500 – 600 ppbv is found in the upper mesosphere around 0.01 hPa (~80 km). There, the highest monthly values are observed in June. In the following months, this anomaly descends and reaches lower levels. In July, the NO_y enhancement of around 10 ppbv reaches the upper stratosphere around 2 hPa, and the increase, although smaller, is visible all the way down to 10 hPa. In the following months, the MIPAS nominal data were unavailable due special observation mode campaigns.

The ALL experiment (Figure 2b) shows a very similar pattern of NO_y as the observations. The NO_y increase of 500 – 600 ppbv in the upper mesosphere around 0.01 hPa is similar as in the MIPAS observations. However, the wintertime NO_y peak below is slightly overestimated in the model compared to MIPAS. This is particularly visible in the lower mesosphere in June, as the modeled 100 ppbv NO_y enhancement reaches 0.1 hPa. The mesospheric anomaly extends into the stratosphere, but remains confined to the upper stratosphere, above 10 hPa, as in observations. The modeled NO_y overestimation suggests that downward transport is somewhat too fast in the model, or the photochemical lifetime of NO_y is too long, or horizontal mixing with mid-latitudes is underestimated. The modeled NO_y enhancement in September stems from a SP event (NOAA, 2018). In contradiction to our results, the EMAC model slightly underestimates NO_y even during polar summer, for two pressure levels, 0.1 and 1 hPa (Matthes et al., 2017). Sinnhuber *et al* (2018) showed similar results in EMAC and KASIMA models and ~~compared~~ overestimation of NO_y in 3dCTM model in southern hemisphere ~~observed~~ compared to ~~by~~ MIPAS ~~with the results of 3dCTM, KASIMA and EMAC chemistry-climate models and also showed overestimation of modeled NO_y~~observations ~~in the southern hemisphere.~~.

The LEE simulation (Figure 2c) shows very similar anomalies as ALL. The largest differences are in the upper mesosphere, where LEE anomalies reach around 400 ppbv, which is underestimated compared to 500-600 ppbv found in MIPAS and ALL. A second interesting difference compared to ALL is the SP event in September. In LEE simulation, it reaches around 60 ppbv, while in ALL it exceeds 100 ppbv. This difference is coming from increased MEE precipitation that coincided with the SP event (see Arsenovic *et al* 2016, Figure 1a). During strong SP events protons can contaminate the highest electron channel, so this channel is excluded from the AIMOS dataset (Yando et al., 2011). Although some degree of contamination is still possible in the lower channels, protons are not the sole cause of the increased NO_y in this SP event. Namely, SP events are often associated to large coronal mass ejections that form a shock in front of them. Once the shock hits the Earth if often leads to a geomagnetic storm which leads to acceleration of electrons of > 30 keV energies. Therefore, increased MEE precipitation often happens very shortly after SP event because the shock and the geomagnetic storm are related to the same coronal mass ejection driver (Asikainen and Ruopsa, 2016).

The MEE simulation (Figure 2d) is drastically different from MIPAS as well as the ALL and LEE simulations. Although NO_y enhancement in the modeled geomagnetically active year exists, it is significantly decreased compared with the previous results. The modeled NOy mesospheric anomaly peak is absent and enhancement of 10 ppbv ~~The pronounced modeled NO_y anomaly maximum from the mesosphere is absent and enhancement of 10 ppbv~~ does not reach the stratosphere. Nevertheless, although less intense, increased NO_y is present throughout the mesosphere and stratosphere, and the NO_y increase in September due to the SP event exceeds again 100 ppbv, as in the ALL simulation.

The reference run in Figure 2e shows NO_y increase due to the SP events in the year 2005. In this year, there were 6 observed SP events in the shown timeframe – May 14, June 16, July 14 and 27, August 22 and September 8 (NOAA, 2018). In the geomagnetically inactive period, 2006-2010, there were no observed SP events in the presented months. Therefore, by excluding electron precipitation, the SP events ~~only~~ alone cannot reproduce the observed features.

From the presented, we conclude that the inclusion of only LEE was sufficient to reproduce most of the NO_y enhancements. The MEE contribution to NO_y increases is minor and brings the model closer to observations mainly in the upper mesosphere. As coronal mass ejections drive SP events and they can have an impact on the precipitation from the outer Van Allen belt

(Asikainen and Ruopsa, 2016; Pierrard and Lopez Rosson, 2016), MEE precipitation could significantly contribute to NO_y increases in such events.

3.2 O_3 anomaly propagation

In the study of Matthes *et al* (2017), ozone responses were evaluated by comparing high and low geomagnetic activity years ~~and not by on/off experiments as done here~~ and their estimate shows good agreement with satellite observations (Fytterer et al., 2015). To evaluate our simulated ozone responses, we follow a similar approach as used in Matthes *et al* (2017), that is, we compared our simulations with observations from MLS. We analyzed the 2005 – 2010 period when both, simulation and MLS data, are available.

Ozone anomalies from MLS observations during the high geomagnetically active year are depicted in Figure 3a. They are calculated as the difference averaged over 70 – 90º S between the active year (2005) and the average of geomagnetically quiescent years (2006 – 2010) divided by the ozone averaged over the whole period (2005 – 2010). Observations show around 20 % less ozone in the upper mesosphere (< 0.1 hPa) occurring mostly in the JJA period. The exception is the SP event on September 8, 2005. It created an ozone anomaly of up to 80 % stretching throughout whole mesosphere. The mesosphere below 0.1 hPa does not show~~s~~ a ~~little~~ statistically significant difference between the geomagnetically active and quiescent years in absence of SP events. The observed negative ozone anomaly appears again around the stratopause in late June and propagates downwards to nearly 10 hPa in early September. The peak ozone anomaly occurs in August around 3 hPa, reaching ~15 %. Our results agree with the results from previous modeling studies (Reddmann et al., 2010; Rozanov et al., 2012; Sinnhuber et al., 2018) and observations (Damiani et al., 2016; Fytterer et al., 2015).

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The ALL simulation (Figure 3b) shows a negative ozone anomaly in the mesosphere as well. However, the magnitude is generally higher (around 30 %) and it is present from May to September. The September 2005 SP event is visible in the model simulations as well and descends from around 1 hPa in late September, reaching 10 hPa in late October. A similar pattern, but less obvious, is seen in the observations. Ozone anomalies in the lower mesosphere (0.5 – 0.1 hPa) are more pronounced in the model than in MLS observations. This is particularly evident in June when the modeled upper-mesosphere anomaly appears to relate to the upper-stratospheric anomaly, in contrast to the observations. This suggest that HO_x production by MEE might be overestimated. In the upper stratosphere model simulations agree well with observations. The decrease propagates downwards, reaching approximately 10 hPa in August, with a peak around 15 % in good agreement with the observations.

Ozone anomalies in the LEE simulation are shown in Figure 3c. Negative ozone anomalies are present mostly in the upper mesosphere (above 0.3 hPa) and have similar magnitude to ALL. Although in the LEE simulation the mesospheric ozone anomaly is overestimated compared to MLS observations, the stratospheric anomaly is almost completely absent. This is surprising, as there are very similar NO_y anomalies in the ALL and LEE simulations (see Figure 2).

Our MEE simulation shows similar ozone anomalies compared to LEE (Figure 3d). The anomalies are confined to a region above 1 hPa and are somewhat reduced compared to LEE and ALL. Similar to LEE, the stratospheric ozone anomaly seen in the observations and ALL simulation is almost absent.

In REF simulation (Figure 3e) most of the ozone anomaly features seen in observations and ALL are missing. The only depletion of ozone in this simulation is caused by SP events in the year 2005. Most of the observed events (May 14, June 16, July 14 and 27, August 22 and September 8) are clearly visible.

A~~R~~ recent study based on CCM WACCM (Andersson et al., 2018) showed ozone anomaly propagation differences between high-A_p and low-A_p winters in the Southern Hemisphere. Their results are comparable with our ALL and LEE simulations. Compared with our ALL simulation, their ozone anomaly in case of all EEP of around 7 % is lower and occurs later (in October as opposed to August). However, their LEE simulation does not show significant ozone anomaly in the stratosphere, which is also the case in our results. In the study of ~~(~~Sinnhuber *et al* (2018) the three analyzed models (3dCTM, KASIMA and EMAC) generally show good agreement with the satellite observations,.

3.3 EEP effect on NO_y, HO_x and O_3

To estimate the total effect of energetic electron precipitation on NO_y, HO_x and ozone, we calculated the differences of experiment simulations (ALL, LEE and MEE) and REF simulation for the geomagnetically active period (2002 – 2005) using the simulated monthly mean values. Note that this is an idealized comparison and it is not directly comparable with observations, as there is always some amount of particle precipitation in the atmosphere (Funke et al 2014), unlike in the LEE, MEE and REF simulations.

The zonal mean of austral winter (JJA) average NO_y differences between ALL and REF is shown in Figure 4a. In polar night, NO_y is transported to lower altitudes by descending air motion. Significant modeled NO_y enhancements are present in the whole mesosphere and upper stratosphere above 10 hPa. Around 0.01 hPa, EPP produced NO_y increases from 50 ppbv at around 60° S, where NO_y lifetime is decreased due to the sunlight, to more than 500 ppbv at the pole, in the polar night. The differences in HO_x between those two experiments are shown on Figure 4b. Increases are mostly confined to the upper mesosphere and they reach the maximum of around 5 ppbv. However, smaller (< 1ppbv) but statistically significant HO_x increase appears in lower mesosphere and upper stratosphere around 60° S. Increases of NO_y and HO_x impact the ozone chemistry. Figure 4c shows changes in ozone concentrations due to electron precipitation. Ozone is significantly reduced throughout the whole polar region above 10 hPa. There are two peaks of ozone anomaly. The maximum decrease of up to 65 % (350 – 400 ppbv) is located in the upper mesosphere. This decrease is more severe than in previous modeling studies (Rozanov et al 2012), but this is because we focus on the geomagnetically active winters, when EPP effects are much more pronounced. The magnitude of ozone depletion is gradually decreasing with height reaching ~15 % (>200 ppbv) at the stratopause. The second ozone depletion peak is located between 10 and 1 hPa, reaching 15 % (>400 ppbv). A similar ozone response as in ALL has been shown by Semeniuk et al. (2011).

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Figure 4d shows the difference between modeled NO_y in LEE and REF simulation. Similarly, as in Figure 2, modeled NO_y in LEE simulation is very similar as in ALL, confirming the fact that the most of the NO_y is coming from LEE. Slight reduction to ALL still exists, visible mostly at 0.1 hPa at 90° S. Here, the value of NO_y is 100 ppbv while it is somewhat more in Figure 4a. A ~~Second~~ second difference is the absence of the enhancement equatorward of 30° S which is present in Figure 4a. Increase of HO_x in case of LEE is illustrated on Figure 4e. Changes of HO_x are very small and statistically insignificant, except for a small (<1 ppbv) increase in the polar upper mesosphere. This is expected as LEE do not produce HO_x. The small increase could be explained by an increase of NO_y which causes small increases of background HO_x through the Verronen and Lehmann (2015) mechanism~~.~~, where ~~As Verronen and Lehmann (2015) pointed out,~~ enhanced NO coming from EEP leads to HO_x repartitioning increasing HO_x concentrations. Figure 4f shows ozone changes due to the LEE. Similar ozone decrease pattern as in Figure 4c exists but with a reduced intensity. The upper-mesospheric reduction reaches 35 % (~200 ppbv) and the upper-stratospheric anomaly is halved compared to ALL (200 ppbv ≙ 10 %). The absence of HO_x increases and reduced ozone anomalies compared to ALL illustrates the importance of MEE.

Figure 4g shows an increase of NO_y due to the MEE. Although MEE cause increases of NO_y, modeled NO_y is significantly reduced in the whole area compared to LEE and ALL simulation. In the upper mesosphere, this increase is around 50 ppbv, or a tenth of the total produced NO_y in ALL simulation. ~~Equatorward f~~Between~~rom~~ 30°-35° S NO_y enhancement is present again, as in ALL simulation. This enhancement is coming from the fact that MEE do not necessarily precipitate inside the polar vortex, as they precipitate in the sub-auroral ovals, which are centered around the geomagnetic pole. In contrast, NO_y coming from LEE descends into the mesosphere in the down-welling air motion inside of the polar vortex. The sum of NO_y increases (not shown) due to the LEE (Fig 4d) and due to the MEE (Fig 4g) closely reassembles NO_y increase as in the ALL case (Fig 4a).

Increases of HO_x due to MEE are presented in Figure 4h. Enhancements are present mostly in the upper mesosphere reaching 4 ppbv. The position and intensity of HO_x is very similar to ALL, but somewhat reduced. Because MEE produce OH, neglecting MEE in climate models would lead to an underestimation of HO_x; neglecting LEE would also lead to an underestimation of HO_x through the changed HO_x partitioning (Verronen and Lehmann, 2015). Changes in ozone concentrations due to MEE are shown in Figure 4i. Negative ozone anomalies are present in the mesosphere and in the upper stratosphere, albeit the stratospheric anomaly is statistically not significant. The ~~Biggest~~ biggest reduction with 35 % (~200 ppbv) is visible in the upper mesosphere. The anomaly in the upper stratosphere (10 – 1 hPa) does not exceed 100 ppbv. Interestingly, summing stratospheric ozone anomaly from LEE (Fig 3f) and from MEE (Fig 3i) does not reproduce ALL ozone anomaly (Fig 3c). The sum of the LEE and MEE ozone anomaly accounts for around 300 ppbv, while ALL shows about 400 ppbv between 10 and 1 hPa. Since the sum of enhanced NO_y due to LEE and MEE corresponds to ALL NO_y and HO_x enhancements occur in mesosphere, this discrepancy in ozone anomaly cannot be chemically explained. It could be caused by changes in dynamics (polar vortex strength) and temperature (which affects reaction rates).

Our results indicate that LEE and MEE are equally responsible for the ozone anomaly in the mesosphere. LEE deplete ozone through the production of large amounts of NO_y, while MEE contribute to the anomaly mostly through production of HO_x, which is the more efficient ozone destructor (Brasseur and Solomon 2005). Both LEE and MEE produce the stratospheric anomaly; however, LEE, through the production of large amounts of NO_y are more important.

4 Conclusions

We used the period 2005-2010 comprising intervals of high and low geomagnetic activity, which is well characterized by stratospheric and mesospheric measurements of NO_y and O_3, to investigate the accuracy of representations of energetic particle forcing in a chemistry-climate model. We assessed the impact of employing a new parameterization of LEE (< 30 keV) recommended for CMIP-6 in combination with the AIMOS parameterization for MEE (30 – 300 keV) on the simulated NO_y, HO_x and ozone variability. We used the SOCOL3-MPIOM climate model and focused on the Southern Hemispheric winter season. We compared NO_y with stratospheric and mesospheric MIPAS observations. The model captures the main features very well, but shows some differences in the winter maxima. LEE can reproduce most of the NO_y features, without including MEE. However, increased MEE precipitation coincident with SP events may be a significant ~~contribute~~ contribution to the ~~reproduce the~~ observed NO_y amounts.

Simulated ozone depletion has been compared to MLS satellite observations, showing that patterns of ozone anomalies during the high EPP year 2005 compared to 2006-2010 match reasonably well. The model overestimates mesospheric ozone anomalies, but in the stratosphere a good match is accomplished. Ozone depletion of up to 15 % is found during July and August and reaches into the lower stratosphere. In essence, without including both LEE and MEE, the stratospheric anomaly cannot be accurately modeled. Future work is required to address the roles of ~~In addition to chemical changes,~~ indirect changes in temperature and dynamics ~~also play a role~~ in the EPP-induced stratospheric ozone variation.

Most of the NO_y in the mesosphere and stratosphere is produced by LEE in the upper mesosphere and lower thermosphere (<0.01 hPa) and transported downwards. A smaller fraction, namely ~10 %, is generated in-situ by ionization due to precipitating electrons of higher energies. These electrons play an important role because they produce HO_x, which depletes ozone near the HO_x source region in the mesosphere. Although not producing HO_x directly, LEE increase NO_y concentrations, which then causes repartitioning of HO_x and an increase HO_x lifetime (Verronen and Lehmann, 2015).

In summary, LEE and MEE lead to a reduction of ozone throughout the mesospheric and stratospheric polar region with a maximum percentage ozone depletion in the mesosphere (-65 %) and a second peak anomaly in the upper stratosphere (-15 %) with respect to the simulation where they are omitted. These chemical EPP signals can cause dynamical changes in the stratosphere that propagate into the lower atmosphere, which eventually affect regional climate (Rozanov *et al* 2012). Therefore, we recommend including both LEE and MEE in climate models.

Author contribution

PA and ER proposed the idea and designed the experiments; PA ~~designed and~~ carried out the simulations and prepared the manuscript. AD analyzed MLS data and made Figure 3. BF provided MIPAS data. TP formulated the general line of research and supervised the project. All authors provided critical feedback and helped shape the research, analysis and manuscript.

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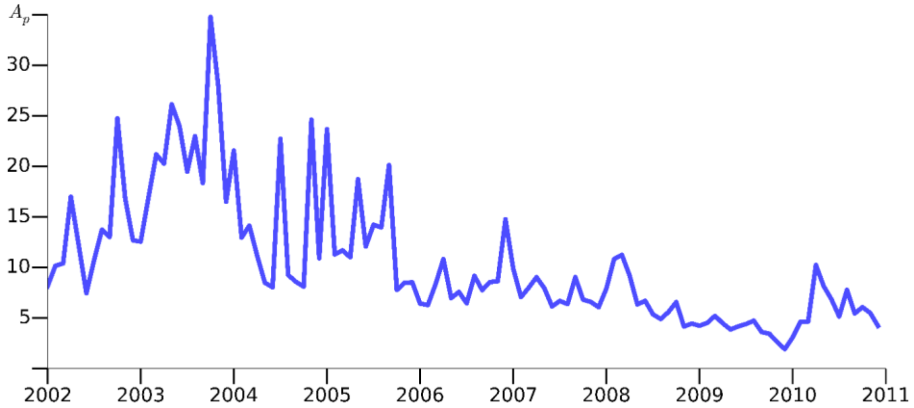

Figure 1: The monthly mean geomagnetic A_p index during the simulated period: Years 2002 – 2005 were rather active, while the period 2006 – 2010 was geomagnetically quiescent (CMIP-6 dataset; Matthes *et al* 2017).

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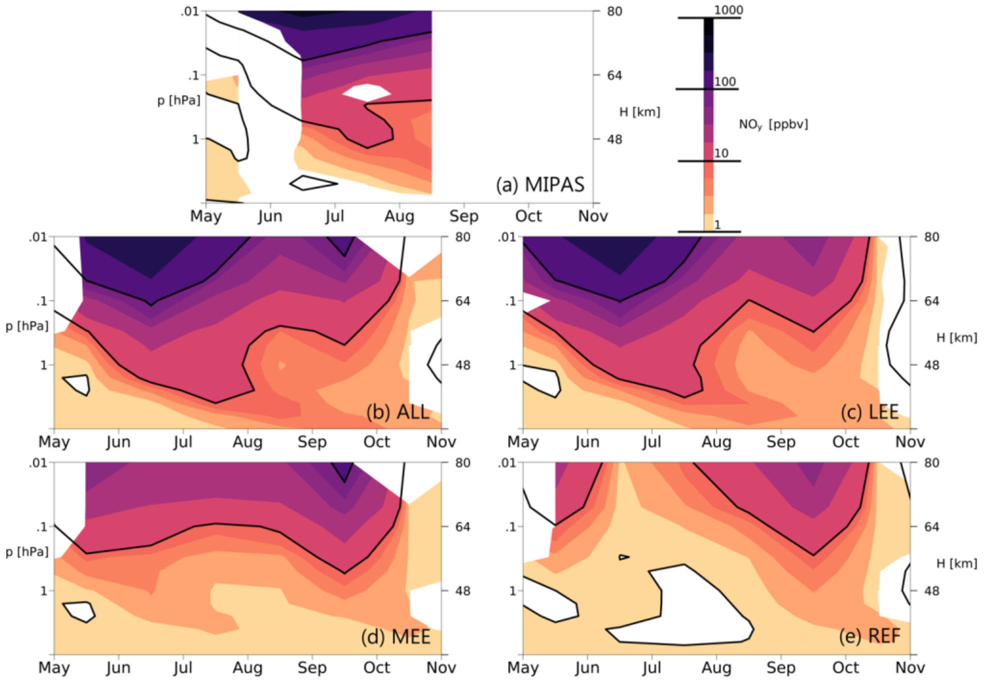

Figure 2: Monthly mean NO$_y$ volume mixing ratio anomaly in ppbv for the Southern Hemisphere (> 70° S average) calculated as difference of the year 2005 and the average of 2006 – 2010. (a) MIPAS observations; (b) ensemble mean of ALL simulations; (c) ensemble mean of LEE simulations; (d) ensemble mean of MEE simulations; (e) ensemble mean of REF simulations. Color levels are 1, 2.5, 5, 7.5, 10, 25, 50, 75, 100, 250, 500, 750 and 1000 ppbv and the ~~Black~~ black contour lines highlight 1, 10, 100 and 1,000 ppbv. Colored regions are significant at the 99 % confidence level (calculated using a Student t-test).

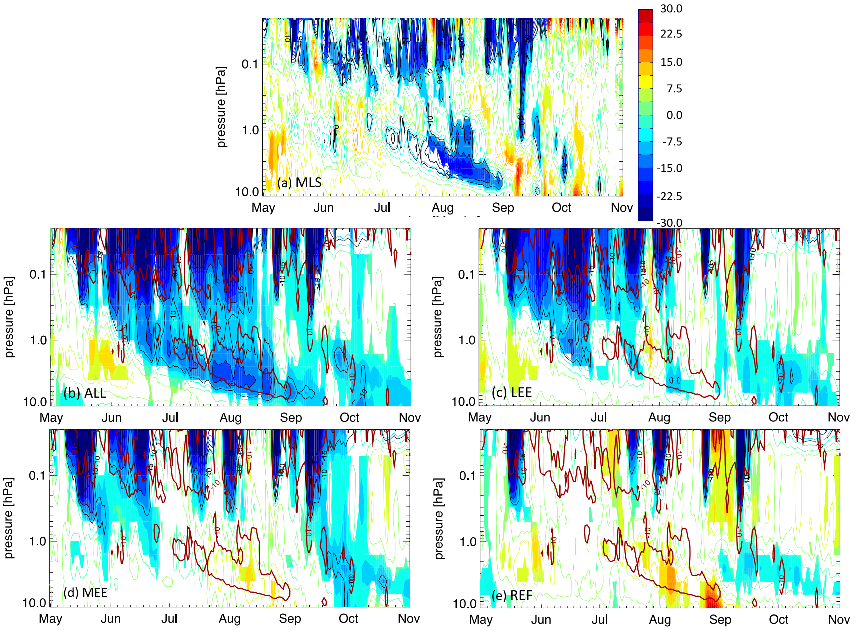

Figure 3: Monthly mean ozone anomaly poleward of 70° S calculated as difference of year 2005 and average of 2006 – 2010 relative to 2005 – 2010 period. (a) MLS observations; (b) ensemble mean of ALL simulations; (c) ensemble mean of LEE simulations; (d) ensemble mean of MEE simulations; (e) ensemble mean of REF simulations. Black lines highlight -10 %, -15% and -50 % and dark red lines mark -10 % from MLS observations on every plot. Note that mesospheric ozone depletion reaches 80-90 % during some strong solar proton events. Colored regions are significant at the 99 % confidence level (calculated using a Student t-test).

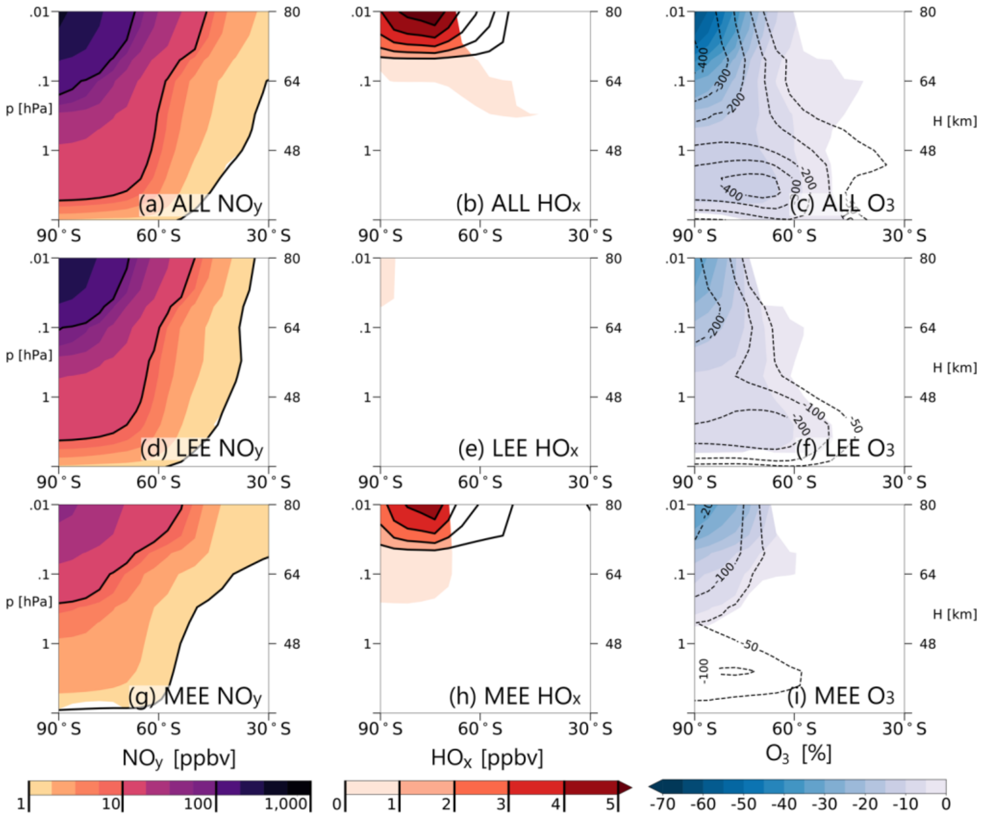

Figure 4: Summary of zonally averaged results. Columns: NO$_y$ (left); HO$_x$ (center); O$_3$ (right). Rows: including ALL energetic particles (top); only with LEE (center); only with MEE (bottom). All panels show results for the geomagnetically active period (2002 – 2005) for austral winter (JJA) from the respective simulations minus the REF simulation. Colors show absolute differences in ppbv for NO$_y$ (color levels are 1, 2.5, 5, 7.5, 10, 25, 50, 75, 100, 250, 500, 750 and 1000 ppbv) and HO$_x$ plots and difference in percent for O$_3$ plots. Isolines show difference in absolute values in ppbv. Colored regions are significant at the 99 % confidence level (calculated using a Student t-test).