Author responses to reviewer comments on paper #acp-2019-1035 ”EPP-NOx in Antarctic springtime stratospheric column: Evidence from observations and influence of the QBO”.

We would like to thank the reviewer for their comments. Our detailed responses are given below.

Reviewer #1 (General comments):

1. The title of the paper suggests that polar springtime EPP-NOx is influenced by the QBO, however, none of the suggested mechanisms results in a modulation of the EPP contribution. Specifically, the authors suggest that (i) the ”amount of the primary NOx source, N2O, transported into the polar regions” is affected by the QBO, and (ii) the ”QBO affects the temperature of the polar vortex and thus the amount of denitrification”. (i) would affect only the background NOx concentration (produced by N2O oxidation) and not the EPP contribution. (ii) would represent a total NOy loss mechanism (independently whether produced by EPP or N2O) and hence would not alter the relative EPP-NOx contribution. In the sense a title like ”Evidence for EPP and QBO modulations of the Antarctic NO2 springtime stratospheric column from OMI observations” would be more appropriate.

   Reply: We would like to thank the reviewer for pointing this out. We very much agree and have changed the title of the paper as suggested.

2. It is suggested that, during eQBO, there is a lack of N2O transported to the polar regions which, in turn, results in a more prominent EPP-NOx contribution and hence better correlation of the observed NO2 column with Ap. This hypothesis is based on Fig 1 of Strahan et al. (2015) indicating a polar springtime N2O depletion during eQBO around 400-600 K (corresponding to approximately 15-25 km) from MLS observations. However, NOy production by N2O oxidation occurs predominantly at higher altitudes (peaking around 30 km which corresponds to a potential temperature level of around 800K) where the MLS observation analysed by Strahan et al. show a N2O increase during eQBO from the equator to around 70S. It is thus more likely that the background NO2 column is enhanced rather than decreased during eQBO because of increased N2O oxidation in the subpolar regions. Note that this is also in consonance with the results shown in Figures 3 and 4.

   Reply: This is correct, our original interpretation of Figure 1 of Strahan et al. (2015) was looking at the wrong altitude range. As pointed out, this is also in line with our results in figures 3 and 4. We have revised all text on this aspect and a grateful for the reviewer on pointing out this.

3. It is further suggested that the ”QBO affects the temperature of the polar vortex and thus the amount of denitrification”, resulting in smaller NO2 losses and hence in- creased NO2 during eQBO. The authors base this explanation on MLS HNO3 observations, indicating an HNO3 increase during eQBO in the 100-10 hPa range. However, it is not clear whether this increase is caused by reduced HNO3 losses (due to a warmer vortex and hence reduced PSC formation) or due to increased productions (e.g. by increased N2O oxidation as mentioned above). In order to proof their ”denitrification” hypothesis, the authors should demonstrate that the HNO3 enhancements during eQBO are linked to temperature increases and/or PSC occurrence. In this context it is worth to mention that the link of PSC coverage and QBO modulation of polar temperature via the Holton-Tan effects is still under debate (see, e.g, Section 4 of Strahan et al., 2015).

   Reply: We have added analysis of MLS temperature observations analogous to the HNO_3 observations. These are presented side by side in Figure 1 here and now included in the manuscript. The temperature analysis suggests that the stratosphere is typically warmer in Aug-Sept in eQBO years. We have revised the text to include the new analysis of the temperature observations to support the HNO_3 analysis and have added information about the temperature observations to the Methods section. We have also included the suggested reference to Strahan et al. on the state of knowledge on this topic in general.

To test whether QBO phase affects denitrification in the Antarctic stratosphere, we analysed

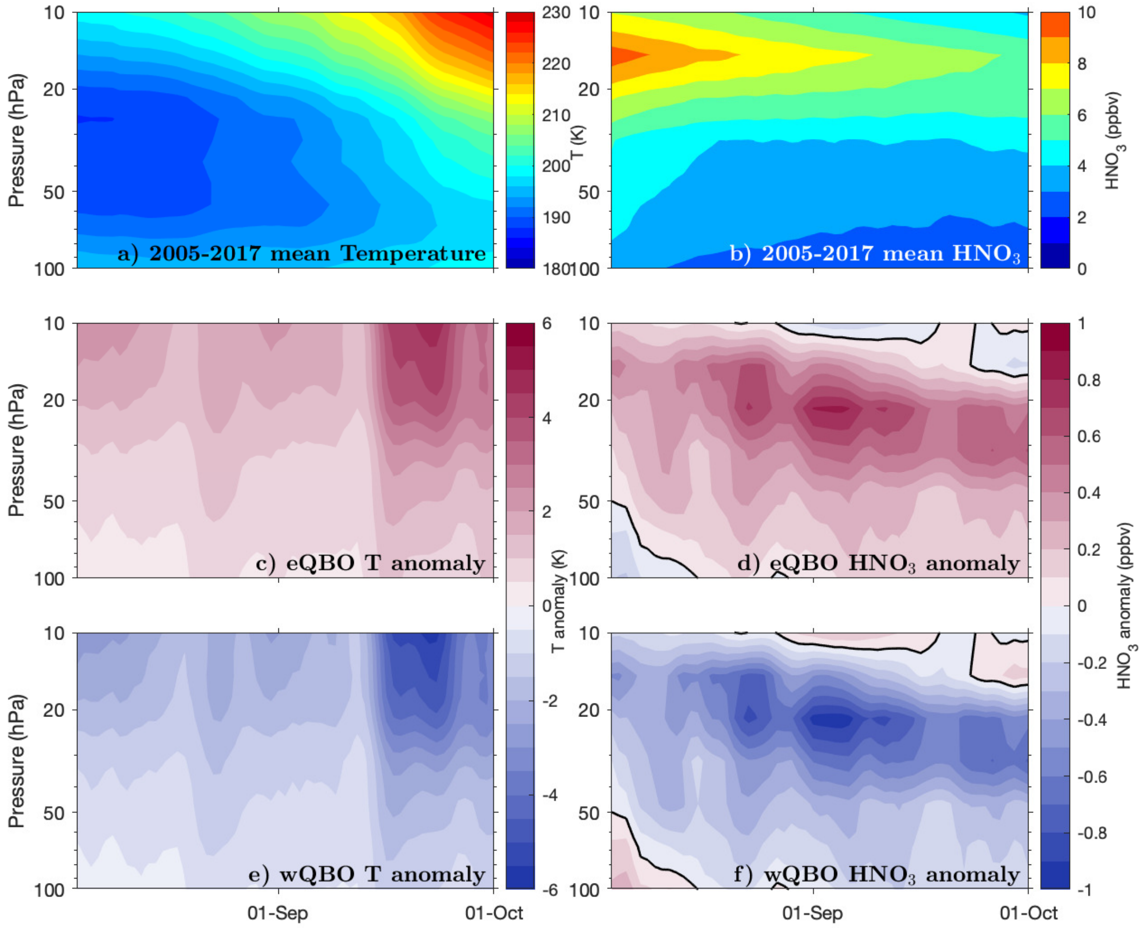

Figure 1: Temperature (left) and HNO_3 (right) mean fields (a-b) for the study period and anomalies for eQBO (c-d) and wQBO (e-f) phases

temperature and HNO_3 *observations from MLS (see section 2.2). Figure 8 a) and b) show the mean temperature and* HNO_3 *respectively, each averaged over 60°S to 82°S for 2005-2017 over the late winter–early spring period, i.e. when the polar vortex is coldest and PSCs are forming. Panels c) and e) show the anomalies from the mean temperature, for eQBO and wQBO years respectively. Panels d) and f) present the anomaly from the mean* HNO_3 *mixing ratio, for eQBO and wQBO years respectively. The vertical pressure range of all panels is 100 hPa to 10 hPa which corresponds to an altitude range of approximately 17 km to 32 km. Figure 8 suggest that eQBO years tend to have more* HNO_3 *(up to 1 ppbv) and higher temperature (up to 4 K) throughout this period, while wQBO years show a consistently negative anomaly in*

*HNO_3 (*down to -1 *ppbv) and lower temperature (down to* -4 *K). Colder temperatures would likely lead to more PSC formation and thus more HNO_3 being removed from the stratosphere (more denitrification) in wQBO years (than in eQBO years). It should, however, be noted here that the link of PSC coverage and QBO modulation of polar temperature via the Holton-Tan effects is still under debate (see, e.g, Strahan et al., 2015).*

Specific comments:

Comment: l23-25: Strahan et al. have shown that the lower stratospheric N2O anomaly at 450 K in the Antarctic polar springtime vortex correlates with the surfzone anomaly at 650 K 12 months earlier, the latter being characterized by enhanced N2O during eQBO.

Reply: We have revised the context of the Strahan et al. work through out our manuscript based on the comments the reviewer has provided above. We now write here:
Recent work by Strahan et al. (2015) has shown that the phase of the QBO influences the transport of N_2O from the surfzone to the polar vortex with a lag of 12 months. Further, their results (Figure 1 of Strahan et al. (2015)) indicate that easterly phase of the QBO during June-July is also generally associated with positive N_2O anomalies in the polar stratosphere between altitudes of ∼24-33 km in September, and opposite for westerly phase of the QBO. Notably for our study, these particular altitudes, at this time, are also affected by large scale transport of mesospheric air masses affected by energetic particle precipitation (Funke et al., 2014a).

Comment: l27: strictly speaking it is HNO3 (not NOx) being removed by denitrification.

Reply: This has been amended
. . . in a process known as denitrification which removes NO_x when it is stored in the HNO_3 reservoir

Comment: l63: the major SSW occurred in January 2004 (not December 2003).

Reply: We have revised this text and it now reads: *However, dynamical effects, driven by the following major sudden stratospheric warming (SSW), indicated that this NO_x was unlikely to have originated from the SPEs. . .*

Comment: l85-86: This sentence is a repetition of what is stated in the preceding paragraph.

Reply: We have removed this as suggested.

Comment: l87 ”...whether this IS detectable....”

Reply: We have corrected this as suggested.

Comment: l147: It is the combined EPP and QBO influence which leads to the most prominent differences between H-Ap/eQBO and L-Ap/wQBO years.

Reply: We have changed the text to reflect this: *the combined influence of QBO and $\hat{A}_p$*

Comment: l186: Figure 5 shows correlations, not NO2 column increases.

Reply: We have revised this sentence to: *The results from Figure 5 suggest that the NO_2 increases at high polar latitudes in September are due to increased EPP/geomagnetic activity, as strong correlations between NO_2 and $\hat{A}_p$ occur in all panels.*

Comment: l204-205: What about wQBO? Fig 7a suggests that correlations improve also for wQBO when considering vortex-only observations.

Reply: We have revised Figure 7 to include the latitude correlations for both eQBO and wQBO years. We have also revised the text accordingly:
Figure 7 c also shows higher correlation with more instances of significance in wQBO years in October than in Figure 5 c though this is more variable than in eQBO years (which is consistent with Figure 5, that wQBO years show lower correlation).

Comment: l206: Consider to add "(see Fig. 7b)"

Reply: We have clarified this as suggested, with the text now reading: *Similarly for the horizontal distribution of the correlations (see Figure 7 b))...*

Comment: l215-220: see general comment (2)

Reply: See response to general comment 2. We have revised this text to clarify how our are results are related to the N2O transport discussed by Strahan et al.

Comment: l223-236: see general comment (3)

Reply: We have revised the text and included analysis of MLS temperature as suggested.

Comment: l229: QBO direction → QBO phase

Reply: We have now corrected this.

Comment: l251: "average rate" implies a time dependence. "average Ap dependence" would be clearer.

Reply: We have revised this as suggested.

Comment: l257: Why should total EPP-NOx only be accounted for in eQBO years?

Reply: This is very true. In writing this we were focused on the larger significances found during eQBO but have now removed this from this text.

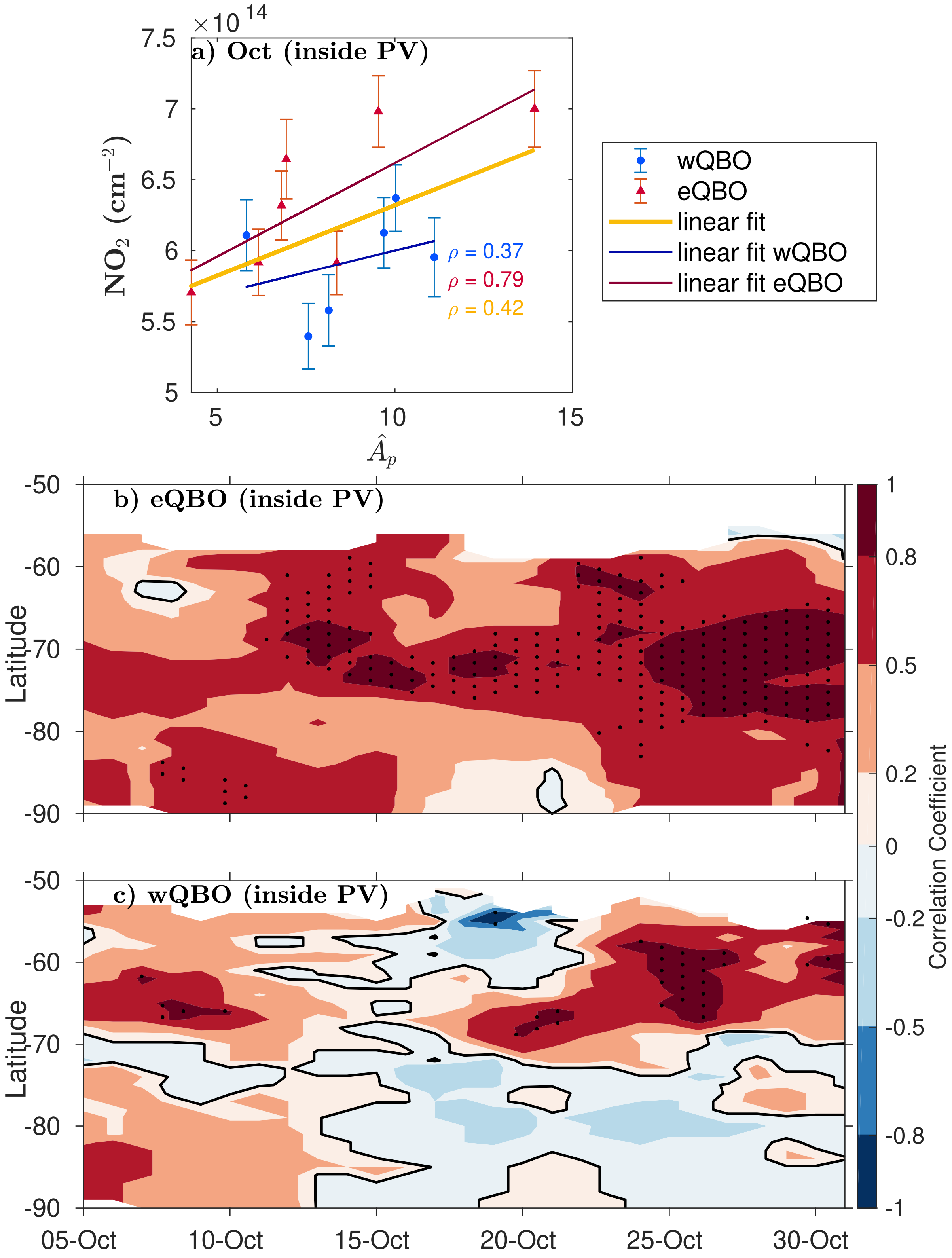

Figure 2: Revised Figure 7, a) now area weighted, b) as before, c) showing the results inside the vortex for wQBO.

Author responses to reviewer comments on paper #acp-2019-1035 ”EPP-NOx in Antarctic springtime stratospheric column: Evidence from observations and influence of the QBO”.

We would like to thank the reviewer for their comments. Our detailed responses are given below.

Reviewer #2 (General comments):

1. Parts of the paper seem to be disconnected and there does not seem to be a clear logical thread to guide the reader. Some of the sections seem to focus on describing figures without explaining why they are relevant to the study and how they relate to the other sections. In particular Sect. 3.1 (esp. Fig. 3) seems unrelated and not relevant to the rest of the paper.

   Reply: We have taken this comment and several of the following comments and added more guidance to the reader, including the later proposed modifications to the introduction (subsections, further information on this work) and result sections. We have also added more clarification on why section 3.1 was included.

2. There seems to be a mixture of terms throughout the manuscript, ”NOx”, ”NOy”, and ”NO2” seem to be used interchangeably. For example the title states ”EPP-NOx” whereas the study focuses on NO2 only. Although according to Brasseur and Solomon, 2005, NO2 makes up around 80% of NOx in the stratosphere, this fact should be noted. The EPP part is not defined at all, Funke et al. 2014a,b use tracer correlations and Randall et al. 2007 use CH4–NO2 correlations to identify *EPP*-NOy, how do the authors discriminate between EPP and non-EPP NO2? A clear definition of these terms and how the authors use them should be given in Sect. 2.

   Reply: This is a very valuable comment. We have changed the title so that it now clearly states that we focus on NO_2. We agree that the terminology is not clear: many of the works looking at EPP impact on atmospheric odd nitrogen focus on the NO_x family, as these gases are mainly available from observations and the immediate increases in odd nitrogen are visible in NO_x. Randall et al. (see e.g. Randall et al. 2006) initially coined the term EPP Indirect Effect (EPP IE) to describe the impact of NO_x produced in the mesosphere by EPP that then descended into the stratosphere. Funke et al. have used a wide range of observations and have been able to investigate the wider NO_y family which includes not only NO_x but its long term reservoirs. For the context of these previous studies and our work both are important, thus we have used both terms NO_x and NO_y as appropriate (e.g. if we refer to a study that analysed NO_y, we use that term). Both have been described in the text and we have now further clarified the use of them across the paper.

   Several works have used tracer correlations to extract solely EPP produced NO_x (or NO_y). Other works have investigated the variability in odd nitrogen resulting in variability in EPP levels (usually proxied by A_p). Our investigation is focused on finding evidence of EPP contribution to column observations, and we unfortunately do not have appropriate tracer information from OMI. Thus we are unable to robustly use the tracer method. To overcome this we performed correlation analysis both for latitudinal coverage and polar average NO_2 observations to find evidence of A_p driven variability in the Antarctic NO_2 column.

   We added the following explanation of the NO_x partitioning to section 2.1 following description of the effective vertical range of the OMI stratospheric NO_2 column:
   At these altitudes, NO_2 makes up about 80% of the total NO_x during daytime (see Brasseur and Solomon, 2005, chapter 5.5). As this corresponds to a large fraction of the total NO_x, we take the OMI NO_2 column measurements to represent a reasonable proxy for the variation in total NO_x.

We also added the following explanation to section 2.3:
Several previous works have used tracer correlations to extract solely EPP produced NO_x *(or* NO_y *when information on the* NO_x *reservoirs is available) (see e.g. Randall et al., 2007; Funke et al., 2014). When tracer information is not available, other works have investigated the variability in odd nitrogen resulting in variability in EPP levels (usually proxied by* A_p*) (see e.g. Seppälä et al., 2007). Here, we focus on finding evidence of EPP contribution to column observations. As we do not have mesosphere-stratosphere descent tracer observations available from OMI, we are unable to use the tracer correlation methods. To overcome this we perform correlation analysis both for latitudinal coverage and polar average* NO_2 *observations to find evidence of* A_p *driven variability in the Antarctic* NO_2 *column.*

3. Do the authors average the 3h Ap or the daily mean Ap? Although other studies use average Ap as well (e.g. Funke et al. 2014a), Ap does not follow a normal distribution and the authors should be aware of that when using the mean as an estimator. This non-normality manifests itself in a very skewed distribution and a large standard devition, particularly the 3h values. Has this been considered in the correlation analysis? I suggest that the authors check that the mean is a valid estimator for the distribution or cite a relevant publication. I also suggest to present the Ap values with error bars in Figs. 2, 4, 7, and A1 and Table 1 (probably based on appropriate quantiles).

   Reply: We use the daily mean Ap values for calculation of our averages. We have also tested calculating median values instead of means, and this leads to the same year-to-year variability shown in Figure 2 which gives confidence that the mean-approach is valid here. We have added error ranges to Table 1 and Figure 2, but we found that the other figures became ”too busy” when more lines were added. We are aware that A_p has a skewed distribution, however, most previous studies also use mean or median values and following the same approach makes our results more comparable.

4. Is the 60°–90° average area weighted? If it is, it should be stated somewhere, Sect. 2 seems the obvious place (l.112). If not, higher latitudes may be artificially amplified in the polar cap average column. And in that case a discussion would be needed to assess the possible differences when taking area weighting into account.

   Reply: Originally the 60°–90° averages were not area weighted. We took this advice and found that area weighting improved our results so we are extremely grateful for pointing this out! Including the area weighting (cos(latitude)), we revised Figures 4 and 7a) and added appropriate description of the weighting method to the text and figure captions. Using identical area weighting to Funke et al. 2014a in Figure A1 also made our results quantitatively more comparable to theirs.

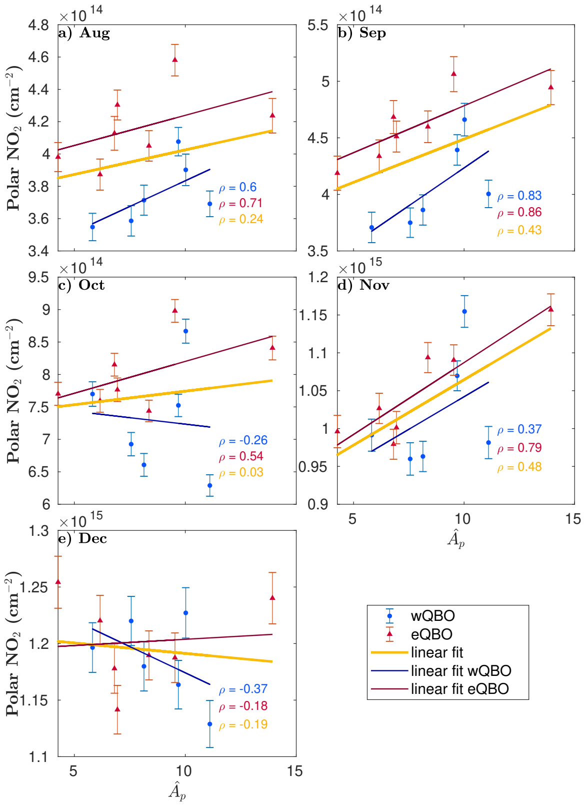

Figure 1: Revised Figure 4, now including area weighting.

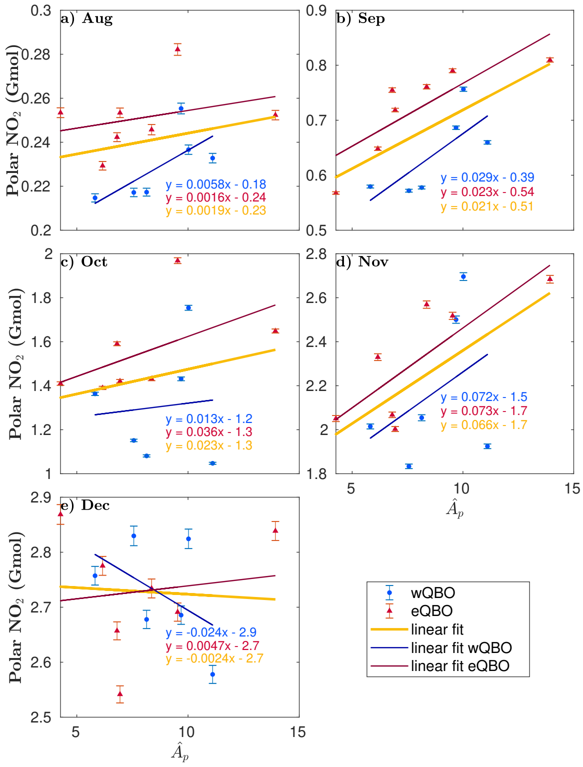

Figure 2: Revised Figure A1, now including area weighting.

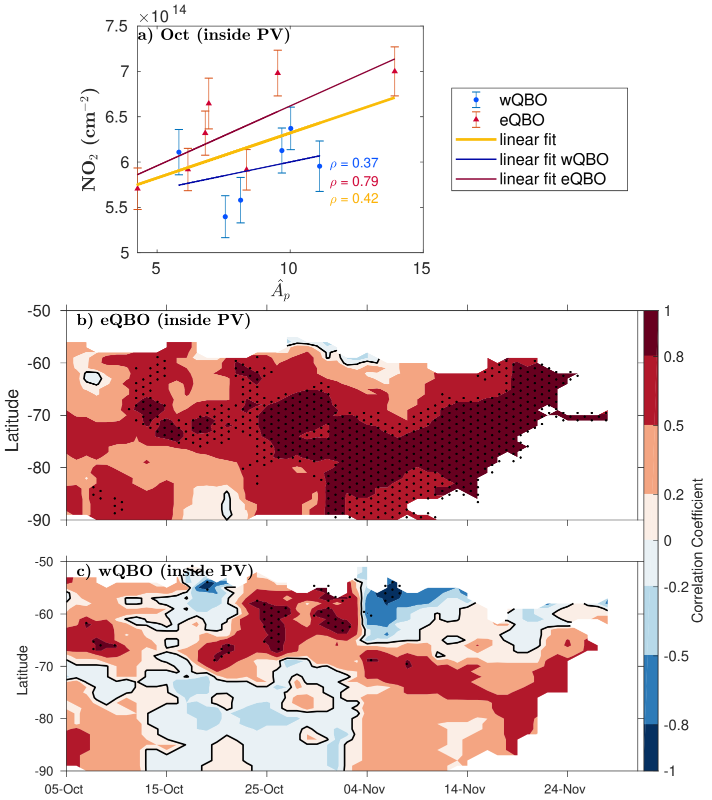

Figure 3: Figure 7 extended through November, a) now area weighted, b) showing results up until the polar vortex break up, c) showing the results inside the vortex for wQBO until the vortex break up.

5. In Sect. 4.1 the authors discuss the possible impact of out-of-vortex air on reducing the correlation between Ap and the NO2 column in October. Why is this presented in the ”Discussion” section and not the ”Results” section? As the authors seem to have an indication about the actual vortex available to them, why isn’t the whole study based on vortex averages instead of whole polar cap averages? That would remove the ambiguity of including non-EPP-NO2 from horizontal transport/mixing in the polar cap average.

   Reply: As suggested, we have moved this section to the Results as recommended.

   Ideally, we would indeed prefer to do the entire analysis for inside vortex air only. However, the method used for locating the polar vortex only works when the ozone hole is present, i.e. mid-September until mid-November. Because of OMI’s observation method and not having sufficiently low ozone amounts in the vortex area before that time, we are not able to do this for the entire time period. We have included here a version of Figure 7 which shows the latitude-resolved correlations throughout November. This shows how the vortex breaks up at this time and thus why we are not able to use the inner polar vortex data for months other than October. We only show October for the monthly mean (panel a)) as it is the only full month that we can calculate for with this method. This panel is now area weighted by cos(latitude). The final version of Figure 7 shows both eQBO and wQBO for the horizontal correlation, but is still only throughout October for the reasons given above

   We have added the following description about the limits of our method:
   Note that we are unable to use this method on months prior to October due to OMI’s viewing method and the ozone column not being sufficiently low to detect a clear vortex edge.

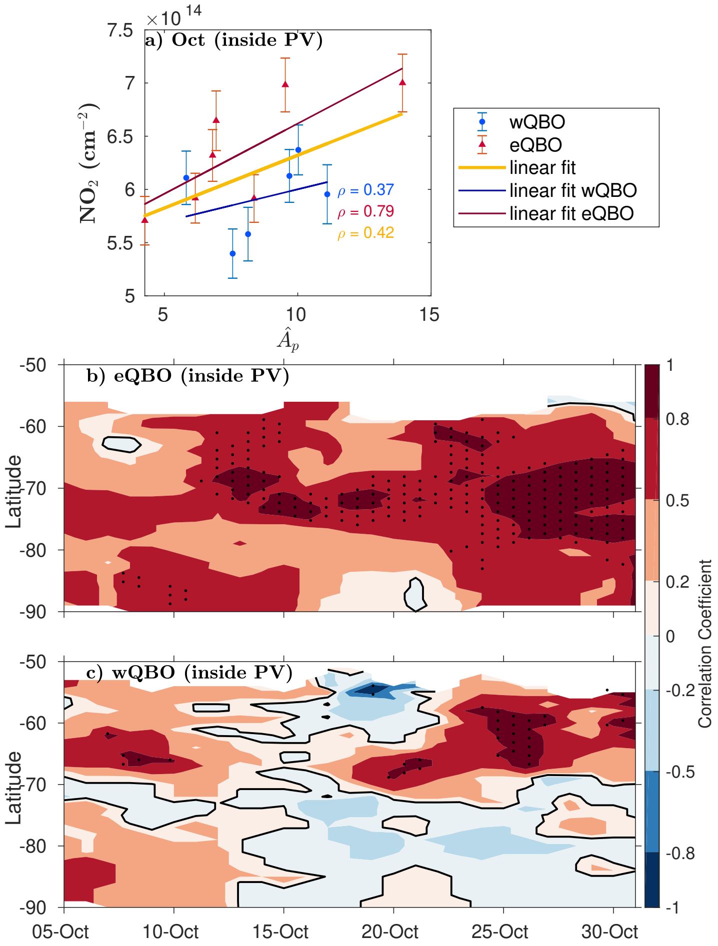

Figure 4: Revised Figure 7, a) now area weighted, b) as before, c) showing the results inside the vortex for wQBO.

Specific comments:

Comment: ll.9–11: Does it really contribute to NO2 or is it just the fraction that changes due to a varying background? I suggest to rephrase these sentences to be clearer, for example how is it linked to the ozone hole? What is cause and what is the effect? See also my other comments below.

Reply: We have revised the text of the abstract to clarify that this NO_x is a significant contributor to the polar NO_2 column when the background changes due to the QBO are accounted for. We have also further indicated the relevance of the ozone hole to our findings, as NO_x catalytically destroys ozone.
Our results suggest that once the background effect of the QBO is accounted for, NO_x produced by EPP significantly contributes to the stratospheric NO_2 column at the time and altitudes when the ozone hole is present in the Antarctic stratosphere. Based on our findings, and the known role of NO_x as catalyst for ozone loss . . .

Comment: I believe the introduction would profit from some additional subsections, e.g.: ll.33–48 ”EPP indirect effect”; ll.49–83 ”Previous work”/”Earlier studies”; ll.84ff ”This work”

Reply: We have divided the introduction text into subsections as suggested.

Comment: ll.85–86: This is a repetition and can be removed.

Reply: We removed the sentence as suggested.

Comment: l.87: A verb is missing: ”... this is detectable ...”

Reply: This has now been corrected.

Comment: The authors do not mention in the introduction that they are going to use (MLS) HNO3 observations, and how they are going to be used. HNO3 is only mentioned in relation to other studies, see also my next point.

Reply: We have added this to the introduction as suggested
QBO conditions influence the probability of PSC formation, and as a result EPP-NO_x may be less/more likely to be removed by denitrification during the winter/early spring during easterly/westerly QBO years. To test for the latter, we will analyse HNO_3 and temperature observations from the Microwave Limb Sounder (MLS), also on-board Aura.

Comment: At the end of the introduction, a guide through the manuscript connecting the parts to the objective raised in the abstract would be helpful, i.e. something like: ”We use the NO2 column data and anomalies correlated to Ap and QBO to assess the impact...” and ”To identify another possible mechanism contributing to the stratospheric EPP-NOx variability, we evaluate MLS HNO3 according to the QBO phase during the same period.”

Reply: This has been amended in the new subsection **This Work** (as suggested above), where we now write:
Here, we use stratospheric NO_2 column observations from the Ozone Monitoring Instrument (OMI) on-board the Aura satellite to investigate EPP as a source of NO_x in the Antarctic later-winter - spring. We have a relatively long satellite period (2005-2017) in which to analyse how this NO_x propagates in the following springtime, and whether this is detectable in the NO_2 column. We also analyse how the phase of the QBO affects the contribution of EPP-NO_2 to the total NO_2 column in springtime. The QBO influence would be likely due to a combination of two effects: 1) QBO phase influences transport of N_2O to the polar region strahan$_e$tal$_2$015$resultinginaincreasedbackgroundNO_x$ source at key times and key

altitudes during easterly QBO years (opposite for westerly QBO); 2) QBO conditions influence the probability of PSC formation, and as a result EPP-NO$_x$ may be less/more likely to be removed by denitrification during the winter/early spring during easterly/westerly QBO years. To test for the latter, we will analyse HNO$_3$ and temperature observations from the Microwave Limb Sounder (MLS), also on-board Aura.

Comment: Observations and methods (Sect. 2): I couldn't find any methods presented here.

Reply: We included a paragraph on how the correlations are calculated as part of this section. However, as this is only one short paragraph, we have revised the section title to "Data sets" as this hopefully better captures the satellite observations, EPP proxy, QBO, and the correlation description.

Comment: Sect. 2.1: are the latitudes geographic or geomagnetic? I assume that the authors refer to geographic latitudes, for completeness, I suggest to state this somewhere in the (sub)section.

Reply: All latitudes are geographic. We have added the following clarification in this section:
Note that all latitudes henceforth are geographic latitudes.

Comment: l.97: I suggest to add some more details about the Aura satellite, such as orbit altitude, inclination, period, and local time.

Reply: We have added the following sentence with a reference to an Aura validation paper:
Aura is in a Sun-synchronous orbit in the "A-train" constellation (orbital altitude of 705 km, inclination of 98°, 16 day repeat cycle), with ascending node crossing the equator approximately at 1:45pm daily (Schoeberl et al., 2008). As a result, OMI measurements take place at the same locations each year.

Comment: ll.102–104: I suggest to use: "The latitudinal coverage is illustrated... . The figure shows ..."

Reply: We have revised this text as suggested.

Comment: ll.105–107: This is a repetition of the earlier statement and can be removed.

Reply: We have removed this text as suggested.

Comment: l.112: Noted in general comment 4, have the measurements been weighted according to their area when calculating the polar cap average (using cos(latitude) for example)? How do the authors account for the lack of measurements north of 70° during Aug–Sep? Has any correction been applied or is it implicitly assumed that the NO2 column is constant (or zero) there? The latter is probably wrong, judging from the curved contours in Fig. 1.

Reply: As addressed in general comment 4 above, we now include area weighting. We do not make any assumptions about the missing values, i.e. we do not assign a value (zero or otherwise) to missing data. We only average the data available.

Comment: Table 1: Please indicate a range for all the values, for example using $\pm(2\times)$ standard error of the mean as in Fig. 4 or appropriate quantiles.

Reply: Error range has been added to $\hat{A}_p$ as $2\times$ standard error of the mean.

Comment: Sect. 2.2: This section appears seemingly without relevance (see comment above about the introduction). It only becomes clear later in Sect. 4.3 when the authors discuss the possible influence of denitrification due to the formation of PSCs. I suggest to better explain how the data are relevant to the study.

Reply: We have revised the text of the introduction to better relate the use of MLS HNO_3 to the study.

QBO conditions influence the probability of PSC formation, and as a result EPP-NO_x may be less/more likely to be removed by denitrification during the winter/early spring during easterly/westerly QBO years. To test for the latter, we will analyse HNO_3 and temperature observations from the Microwave Limb Sounder (MLS), also on-board Aura.

Comment: l.116: Geographic or geomagnetic latitudes? I suggest to state that somewhere at the beginning.

Reply: This has been addressed in an earlier comment on section 2.1.

Comment: Sect. 2.3: Mentioned in general comment 3, Ap has a non-normal distribution, how do the authors deal with that?

Reply: This has been addressed in general comment 3 above.

Comment: l.122: the reference should be probably to Funke et al., 2014a instead of b.

Reply: This is has been corrected and now reads Funke et al., 2014a.

Comment: l.132: How was the confidence interval estimated?

Reply: The Spearman rank correlation calculation using Matlab's statistics package returns the corresponding p-values for testing the null hypothesis. We determine statistical significance using these p-values. We now write in the text:

Statistical significance is here defined as correlations significant at $\geq$95% (i.e. p-value of ≤ 0.05).

Comment: Sect. 2.4: Strahan et al., 2015 use a different definition of QBO which results in a different division of eQBO and wQBO years compared to the one presented here. The authors should comment on that and how it would influence the results (see also below).

Reply: The applicability of Strahan et al. 2015 is now addressed in Sec 4.2. Although their designation of QBO phase differs slightly from ours, this will not largely affect their comparability, as our designations only differ for one year of a 10 year study.

Comment: ll.136–137: This sentence is confusing, "take" does not seem to be appropriate here, please rephrase.

Reply: This sentence has been rephrased to:

We designate years when the zonal mean zonal wind direction is easterly as easterly QBO.

Comment: Fig. 2: Error bars and a $\hat{A}_p = 8.5$ line would be helpful to visualize the Ap ranges and the division into low and high Ap years (only needed if Sect. 3.1 is kept in the manuscript, see below).

Reply: The value 8.5 was a typo in the previous version of the manuscript, 8.3 being the correct average $\hat{A}_p$. We have revised all instances of this. We have also revised the figure to include errorbars and a line as suggested. The caption has also been revised accordingly.

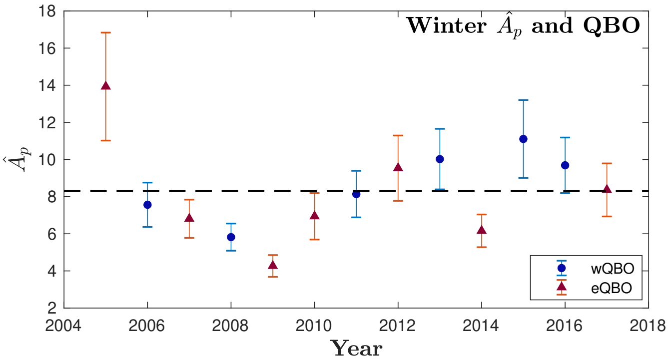

Figure 5: Revised Figure 2, now including horizontal line at $\hat{A}_p$=8.3 and error bars on the $\hat{A}_p$ values.

Comment: Results (Sect. 3): A little guide through the results would be helpful, as in "We investigate anomalies to assess ...", "Then, polar averages are correlated in order to ...", "Latitudinal correlations are used to ..."; either at the beginning of Sect. 3 or at the beginning of the respective subsections.

Reply: A motivation for each figure has been included at the start of each subsection:
- We first investigate the anomaly from the mean for each of the four different categories of this study, eQBO H-$\hat{A}_p$, eQBO L-$\hat{A}_p$, wQBO H-$\hat{A}_p$, and wQBO L-$\hat{A}_p$. This is to show how NO_2 column evolves in the springtime in the different conditions and to further justify the splitting of years based on QBO phase.
- This is to investigate whether there is a relationship $\hat{A}_p$ size and NO_2 column, and how this is affected by QBO phase.
- This shows how correlation between $\hat{A}_p$ and NO_2 column evolves over time and latitude, and different QBO phase.

Comment: Sect. 3.1: As mentioned in general comment 1, this section does not seem to play a role in the rest of the manuscript and raises a lot of questions. For example, I count only two years (2005 and 2012) for panel (a), five (2007, 2009, 2010, 2014, and 2017) for panel (b), and three each for (c) and (d). How robust are those means then? How does it vary with the choice of QBO definition? Strahan et al. 2015 list 2011 as eQBO, not wQBO, how does that affect the results? How robust are the results with respect to the Ap distribution? 2017 for example could also be a high-Ap year (it is close), how would that change Fig. 3?

Reply: As stated above, the earlier value of 8.5 was a typo. Our results in panel a) were actually for 2005, 2012 and 2017, and b) were 2007, 2009, 2010 and 2014. This figure is included to give the reader background on the background behaviour of NO_2 under each of the different conditions. It is intended to provide further justification for the methods used in the study (i.e. that both $\hat{A}_p$ and QBO affect seasonal evolution of column NO_2).

Comment: l.142: "... the mean deducted ..." What mean? The mean as shown in Fig. 1? If yes, please refer to that figure.

Reply: Yes it is the mean in Figure 1, we have clarified the text accordingly: Figure 3 presents the average anomaly, (i.e. the mean as in Figure 1 deducted).

Comment: However, I suggest to remove that section entirely and to start the results with the scatter plots in Sect. 3.2. The split into high and low Ap is not used later, the authors then only divide into eQBO and wQBO years.

Reply: The justification for this figure has been clarified.
We first investigate the anomaly from the mean for each of the four different categories of this study, eQBO H-$\hat{A}_p$, eQBO L-$\hat{A}_p$, wQBO H-$\hat{A}_p$, and wQBO L-$\hat{A}_p$. This is to show how NO_2 column evolves in the springtime in the different conditions and to further justify the splitting of years based on QBO phase.

Comment: Sect. 3.2: Fig. 4 caption: "The yellow line ..."

Reply: This has been added.

Comment: l.151: Again, please indicate if the data have been weighted by the area. It is only needed once, though. And again, what about the missing data in Aug–Sep?

Reply: This comment has been addressed above.

Comment: l.153: How was the linear fit achieved? Were the data weighted by their uncertainties or not? What about uncertainties in $\hat{A}_p$*? Please be more specific here, in particular since this is later related to Funke et al., 2014a.*

Reply: We use least squares linear fitting. This was mentioned in section 3.2 but we have added the information to the figure captions as well. We do not apply weighting as the errors in the NO_2 *columns remain fairly consistent from year to year (see Figure 1 here). Note that the means are based on around* 2×10^5 *daily observations for each monthly mean.*

Comment: l.156: "... [not] fully encompass the entire polar region ..." How do the authors deal with it? Are the averages calculated only up to 70° in those cases? Is the missing area filled with a constant value or even with zeros? I suggest to clarify these points.

Reply: As explained above, the averages are calculated for the actual data available. The missing area is treated as missing values and thus does not contribute to the mean. This has been further clarified in the text:
The missing data in August and September is treated as missing values in the mean calculation and as such does not contribute to the mean in these figures.

Comment: l.162: "... have consistently lower NO2 column values, especially in August–September." May this be the result of omitting higher latitudes or implicitly replacing them by a constant or even zero? What about the influence of area (cos(latitude)) weighting?

Reply: As stated earlier we did not apply area weighting (but do now, thank you for the suggestion). We do not assign values to missing values but rather omit them from calculations.

Comment: ll.163–174: Related to my general comment 2, how do the authors define the EPP part of the measured NO2 columns? Why is Fig. A1 put into a non-existing appendix and not included here? I suggest to move that figure here as Fig. 5. Why not use the same Ap weighting scheme as described in Funke et al., 2014a? Note that they used that procedure for a reason and it would make the two studies really comparable on an absolute scale.

Reply: Some of the aspect of this comment have been addressed in the earlier responses. The Ap weighting factors of Funke et al., 2014a are based on their linear regression of MIPAS EPP-NO_y *data and thus, unfortunately, not possible for us reproduce - we do not have tracer data or vertically resolved* NO_y *needed for their sophisticated approach. Since the simple average* A_p *approach has been found a good proxy for EPP by many previous studies, we rely on that here as well. With figure A1 we are concerned that it is too similar to figure 4 to be included in the main text. This was also advise from more senior colleagues. If moving the figure is supported by the ACP handling editor we would be very happy to move the figure to the main text.*

Comment: Sect. 3.3: l.182: Again, what part of the OMI NO2 column is EPP-NOx here?

Reply: We have added information of the NO_2 *contribution to the total* NO_x *budget at the OMI effective altitudes (as discussed above). Since* NO_2 *makes up about 80 % of the total* NO_x *and we find significant evidence of high positive correlation between this* NO_2 *column and* A_p *this indicates that the EPP-*NO_x *reported by others in past studies does indeed show up in the spring time column. We have added various clarifications to several parts of the paper that we think now clarifies these points more.*

Comment: l.185: How was the significance determined? Similar in caption of Fig. 5.

Reply: We have added more information to earlier section 2 and this text has been clarified:
Stippling indicates correlation significant at $\geq$95% level.

Comment: l.186: I suggest to replace "from Fig. 5" by "shown in Fig. 5".

Reply: This has been revised as suggested

Comment: Discussion (Sect. 4): l.192: I suggest to add an article "... presented in the previous sections ..."

Reply: This has been revised.

Comment: l.192 cntd.: "less significant" than what? Using the frequentist language as in the other parts of the manuscript, the results are either significant or not (according to the chosen significance level). Do the authors mean "less correlated" (ρ is around zero)? Or: "[the correlations] ... are less clear/smaller/weaker"?

Reply: We have revised this to:

were found to have fewer occurrences of statistical significance in October than the surrounding months.

Comment: l.195: I suggest to remove "the month of".

Reply: This has been removed as suggested.

Comment: ll.196–211: As suggested in general comment 5, the study could be based on the polar vortex averages instead of the polar cap mean. I also suggest to move this part to the results, not the discussion.

Reply: This has been moved to the results section as suggested. We cannot base this study on polar vortex averages as our method for isolating the vortex is not valid for the entire winter-spring period, see response to general comment 5.

Comment: l.205: What about the vortex shape variability in other months?

Reply: See response to general comment 5 on why the only full month this method is applicable for is October.

Comment: ll.208-211: I couldn't make any sense of that rather convoluted sentence, I suggest to rephrase it to be clearer; "thus" seems to be the wrong word here.

Reply: This sentence has been revised to:

The reappearance of correlations in eQBO years in November in Figure 5b) is likely a mixing effect, with the break down of the polar vortex around this time leading to vortex air being mixed with extra-vortex air resulting in the NO_2 distribution not being as skewed as it was when contained in the vortex.

Comment: l.212: The word "now" seems to be misused, I suggest to use "in our study".

Reply: We have revised this to: has generally been lower in the past decade *to be more specific about the time period.*

Comment: ll.213–214: Leaving the complications with Ap aside, the implication is only valid if the authors have a particular model/mechanism in mind that "generates a proportional response". Without that model or mechanism, the results merely suggest this response. I recommend to soften the wording accordingly, or to present a clear mechanism that links cause and effect.

Reply: We have removed this sentence.

Comment: Fig. 6: Is this the October OMI ozone average column using all years? Or just one example month? What about the year-to-year variability of the vortex shape?

Reply: We use co-located ozone observations for each individual NO$_2$ observation to determine the location of the polar vortex edge. Figure 6 is an example of one day, 19th October 2014. This is repeated every day in October in the study period to find the daily polar vortex extent. We then apply this to NO$_2$ to isolate the in-vortex NO$_2$ every day. We have revised both the text and the figure caption to make this clear.

We perform this method for every day of October in the study period to find the daily vortex extent. This is then used to locate NO$_2$ that is inside the vortex for every day in October over the study period. An example of how the ozone column and the estimated vortex edge are reflected on the NO$_2$ column measurement for one day of the study (19th October 2014) is shown in Figure 6.

The caption now reads:

Vortex edge identification based on OMI ozone column for the 19th October 2014 . . . This method is repeated for every day in October throughout the study period.

Comment: Sect. 4.2: Since this is the "discussion" section, the influence of the different QBO definitions should be discussed. The decreased N2O concentrations were observed in the average eQBO according to their (Strahan et al., 2015) definition of QBO (which is different from the one used here). Similarly, the mechanism that connects N2O and NO2 could be repeated to make clear why the Strahan et al., 2015 study is relevant here.

Reply: We have revised the text to address the differences in QBO defintion: Although their designation of QBO phase differs slightly from ours, this will not largely affect their comparability, as our designations only differ for one year of a 10 year study. *We have repeated the mechanism connecting N2O and NO2 and clarified the relevance of the Strahan et al. study.*

Comment: l.216: I suggest to swap "the" and "that".

Reply: This has now been corrected.

Comment: l.218: I suggest to remove "clearly".

Reply: We have revised this as suggested.

Comment: l.220: I suggest to replace "more" by "a larger fraction".

Reply: We have revised this as suggested.

Comment: Sect. 4.3: Fig. 8: The panels are missing the (a), (b), and (c) indicators to be consistent with the figure caption. Caption (b): "anomaly from the mean", I assume the 3-day mean as shown in panel (a) is subtracted, please clarify that.

Reply: This figure has been revised, with appropriate labels included. We have also updated the caption.

Comment: l.222: I don't understand this sentence, what is meant by "the affected transport"? I suggest to rephrase that sentence to be clearer, and to remove "obviously" from it.

Reply: We have revised this sentence to clarify the motivation behind this part of the discussion.

Here we discuss reasons for the consistently lower amounts of NO$_2$ in wQBO years that we pointed out in Figure 4. We suggest that this is due to the effect of denitrification in the polar region.

Comment: l.224: An article seems to be missing: "A colder polar vortex ..."

Reply: We have now corrected this.

Comment: l.225: "As discussed earlier", where? A reference to the relevant section would be helpful.

Reply: We have revised the text to:
As discussed in the Introduction, PSCs affect the heterogeneous chemistry in the polar region...

Comment: l.226–228: This sentence is hard to understand, I suggest to rewrite it, for example using Thus or Therefore instead of "So".

Reply: We rewrote the sentence and it now reads:
Thus, for years with more PSCs (i.e. wQBO) more denitrification would likely occur, resulting in the depleted NO_2 column reported here. This could also explaining the lower incidence of significant correlation in the wQBO cases.

Comment: l.234: I suggest to use "down to -1 ppbv" and to remove "clearly".

Reply: We have revised the text as suggested.

Comment: ll.235–236: If PSCs are really responsible for the loss of HNO3 due to denitrification, have the authors considered additional observations of e.g. PSC fraction or temperatures during eQBO or wQBO that would support that mechanism? I suggest to include a short comment or reference.

Reply: We have revised this section to include analysis of MLS temperature observations in the polar lower stratosphere in the springtime, comparing the different QBO phases. The vortex is colder in wQBO years, supporting our hypothesis that PSCs are more likely to occur in wQBO years.

Comment: Conclusions (Sect. 5): ll.244–248: I suggest to move that part or a some version of it to the discussion section as it summarizes the assumed mechanisms. It would also fit at the end of the introduction to help the reader to understand the purpose of the study.

Reply: We took the advice and moved this section to the introduction as suggested. The end of the Introduction now reads:
The QBO influence could be likely due to a combination of two effects: 1) QBO phase influences transport of N_2O to the polar region strahan$_{e}tal_{2}015resultinginaincreasedbackgroundNO_{x}$ source at key times and key altitudes during easterly QBO years (opposite for westerly QBO); 2) QBO conditions influence the probability of PSC formation, and as a result EPP-NO_x may be less/more likely to be removed by denitrification during the winter/early spring during easterly/westerly QBO years. To test for the latter, we will analyse HNO_3 and temperature observations from the Microwave Limb Sounder (MLS), also on-board Aura.

Comment: l.252: This is a confusing sentence, how does the ozone hole suddenly come into play?

Reply: We agree with the reviewer. Our aim was to highlight the potentially changing role of solar activity via EPP as a modulation source of polar NO_x as the stratospheric chlorine loading is changing. We have revised the text and it now reads:
We present evidence of contribution from EPP-NO_x in the Antarctic stratosphere at a time when *halogen activate ozone loss is taking place*.

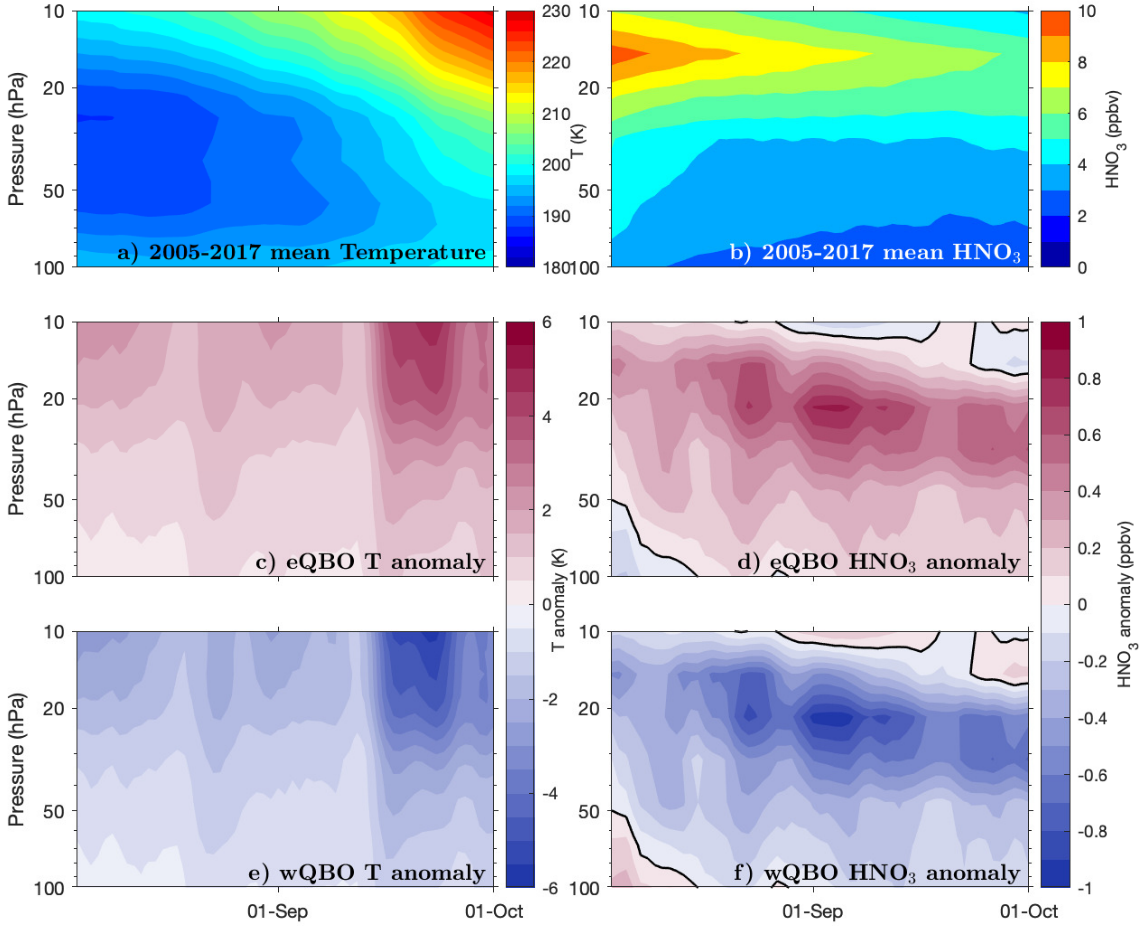

Figure 6: Temperature (left) and HNO$_3$ (right) mean fields (a-b) for the study period and anomalies for eQBO (c-d) and wQBO (e-f) phases

Comment: ll.256–259: This conclusion is stretching it a bit too far in my opinion. According to the presented study, the EPP-NOx (in form of NO2) does not change with QBO phase. Instead, the background NO2 changes due to source and sink changes. As a consequence, the fraction of EPP-NOx (NO2) on the overall amount varies with QBO phase. The authors may consider rephrasing their last conclusion a bit, such that the larger EPP-NOx fraction may need to be considered when considering the net effect of NO2 on ozone chemistry (resp. recovery).

Reply: We have revised this part according to the suggestion and it now reads:
Our results suggest that, as chlorine activation continues to decrease in the Antarctic stratosphere following the Montreal Protocol (Solomon et al., 2016), the total EPP-NO$_x$ *(in addition to SPEs as pointed out by Stone et al., 2018) should be accounted for in predictions of Antarctic springtime ozone recovery. Future studies should investigate the effects the larger EPP-NO*$_x$ *fraction when investigating the net effect of NO*$_2$ *on the fragile ozone chemistry in the springtime.*

Comment: References: There are two Seppälä et al., 2007 references listed, they should be separated with (a) and (b). They are referenced in ll.34 and 76 at least, which is which?

Reply: This was an unfortunate issue with LaTeXand BiBTeX, which we used for compiling the bibliography. This has now been corrected with ”a” and ”b” added as appropriate.

~~EPP-NO$_x$ in~~ Evidence for EPP and QBO modulations of the Antarctic NO$_2$ springtime stratospheric column ~~: Evidence~~ from OMI observations~~and influence of the QBO~~

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Abstract. Observations from the Ozone Monitoring Instrument (OMI) on the Aura satellite are used to study the effect of energetic particle precipitation (EPP, as proxied by the geomagnetic activity index A$_p$) on the Antarctic stratospheric NO$_2$ column in late winter-spring (Aug-Dec) during the years 2005–2017. We show that the polar (60°S-90°S) stratospheric NO$_2$ column is significantly correlated with EPP throughout the Antarctic spring, until the breakdown of the polar vortex in November. The strongest correlation takes place during years with easterly phase of the quasi-biennial oscillation (QBO). We propose that the QBO likely affects the polar springtime ~~EPP-NO~~NO$_x$ in two ways: firstly by ~~modulating the~~ the known modulation of the amount of the primary NO$_x$ source, N$_2$O, transported to the polar region ~~. Secondly, the QBO affects the~~ and secondly, by influencing the temperature of the polar vortex and thus the amount of denitrification occurring in the polar vortex~~, also verified from~~. The latter is supported by temperature and HNO$_3$ observations from the Microwave Limb Sounder (MLS/Aura). Our results suggest that once the background effect of the QBO is accounted for, NO$_x$ produced by EPP significantly contributes to the stratospheric NO$_2$ column at the time and altitudes when the ozone hole is present in the Antarctic stratosphere. Based on our findings, ~~we recommend~~ and the known role of NO$_x$ as catalyst for ozone loss, we propose that as chlorine activation continues to decrease in the Antarctic stratosphere, the total EPP-NO$_x$ ~~should~~ needs be accounted for in predictions of Antarctic ozone recovery.

1 Introduction

In the polar stratosphere, the dominant source of odd nitrogen, NO$_x$ (NO + NO$_2$), is produced via the oxidation of nitrous oxide, N$_2$O (Brasseur and Solomon, 2005):

$$\mathrm{N_2O + O(^1D) \longrightarrow 2NO.} \tag{1}$$

This reaction requires the presence of excited oxygen atoms O(^{1}D), which are produced in the atmosphere by photolysis of ozone (O$_3$) and thus depend on sunlight being present. As a result, NO production via reaction (1) only takes place outside polar winter conditions. Following reaction (1) the existing NO can be converted to NO_2 in reaction with ozone:

$$NO + O_3 \longrightarrow NO_2 + O_2. \tag{2}$$

As N_2O production *in situ* in the polar stratosphere is insignificant, the polar stratospheric NO_x production is highly dependant on the amount of N_2O transported from the tropics (Brasseur and Solomon, 2005). This principal source of polar N_2O is injected from the troposphere into the stratosphere at equatorial latitudes. It is then transported towards the polar regions by the large scale Brewer-Dobson circulation. ~~The strength of the Brewer-Dobson circulation is modulated by the Quasi-Biennial Oscillation (QBO): Strahan et al. (2015) have~~ Recent work by Strahan et al. (2015) has shown that the ~~easterly/westerly~~ phase of the QBO ~~(at ~20 hPa) during the Southern Hemisphere (SH) winter results in anomalously low/high Antarctic polar stratospheric~~ influences the transport of N_2O from the surfzone to the polar vortex with a lag of 12 months. Further, their results (Figure 1 of Strahan et al. (2015)) indicate that easterly phase of the QBO during June-July is also generally associated with positive N_2O ~~in the following spring (September).~~ anomalies in the polar stratosphere between altitudes of ~24-33 km in September, and opposite for westerly phase of the QBO. Notably for our study, these particular altitudes, at this time, are also affected by large scale transport of mesospheric air masses affected by energetic particle precipitation (Funke et al., 2014a).

The main pathway to NO_x loss is via photolysis (Brasseur and Solomon, 2005). During polar night conditions when little to no sunlight is available, this results in long chemical lifetime (weeks to months) for the NO_x family. However, NO_x can be removed from the lower stratosphere during the polar night in a process known as denitrification which removes NO_x when it is stored in the HNO_3 reservoir. This requires the winter vortex to be cold enough that polar stratospheric clouds (PSCs) form. Denitrification occurs when reactive nitrogen (particularly NO_2) is converted into HNO_3 in the lower stratosphere (Santee et al., 1995). HNO_3 is readily incorporated into PSCs, removing gaseous HNO_3 from the lower stratosphere as it eventually falls into the troposphere via gravitational sedimentation (Brasseur and Solomon, 2005).

1.1 EPP indirect effect

It is now well established that precipitating energetic particles can drive large enhancements in NO_x quantities in the polar atmosphere (see e.g. Seppälä et al., 2007a; Funke et al., 2014a, b). Energetic particle precipitation (EPP) is the flux of charged particles (protons and electrons) of solar and magnetospheric origin into the Earth's atmosphere. The charged particles are guided to the polar regions by the Earth's magnetic field. Once reaching the atmosphere, they ionise the main neutral gases, N_2 and O_2. The chain of ion-neutral reactions that follows the ionisation then leads to increases in NO_x species (this is known as "EPP-NO_x"), particularly in the mesosphere and lower thermosphere (Brasseur and Solomon, 2005). EPP manifests as energetic electron precipitation (EEP) as well as proton precipitation, which in the form of solar proton events (SPEs) is the most extreme from of EPP (see e.g. Seppälä et al., 2014). SPEs are usually associated with coronal mass ejections (CMEs) and thus, while the particles are highly energetic and have the ability to ionise as far down as the stratosphere, the events are short (hours to days) in duration, and occur sporadically. Conversely, EEP is always present in some form and is mostly dependant on solar wind speed (Funke et al., 2014b). Due to the lower energies of the electrons, EEP driven *in situ* NO_x increases typically occur in the mesosphere and above (Turunen et al., 2009). When EPP occurs over the winter pole, the mesospheric NO_x has a long chemical lifetime and can be transported downwards into the stratosphere inside the polar vortex. Once in the stratosphere, these NO_x enhancements are effective at catalytically destroying ozone (see e.g., Jackman et al., 2008, and references therein). As NO_x is not formed from N_2O during winter, EPP becomes a significant contributor to the polar winter NO_x budget (Funke et al., 2014b).

1.2 Previous work

Several previous studies have examined the effects of EPP on polar winter NO_x, or the wider NO_y family which includes both NO_x and its reservoir species such as NO_3, N_2O_5, HNO_3, and $ClONO_2$. We will summarise the findings of the key observational works, with particular focus on those with SH or NO_x transport aspects, in the following. Note that we will use the the term NO_x or NO_y depending on which the study in question addressed.

Randall et al. (1998) reported stratospheric NO_2 observations from the Polar Ozone and Aerosol Measurement (POAM II) over three polar late-winter/early-spring periods. They found evidence to suggest that NO_x from the SH polar mesosphere was transported down into the stratosphere inside the polar vortex during the winter. They also suggested that the observed enhanced levels of stratospheric NO_x in 1994 could be, at least partially, due to production by EPP that took place at higher altitudes before the downwards transport. Based on their analysis, Randall et al. (1998) suggested that NO_x transported to stratosphere from the mesosphere and above during the polar winter should be observable in Antarctic NO_2 column measurements.

Siskind et al. (2000) used Halogen Occultation Experiment (HALOE) observations from the UARS satellite between 1991-1996 to track NO_x enhancements in October in the SH polar region. They found that the year-to-year variability in NO_x inside the polar vortex followed variability in wintertime mean auroral A_p index, a measure now frequently used for overall EPP levels (Matthes et al., 2017). At the time, they found that the peak NO_x enhancements from "auroral" activity corresponded to around 3-5% of the total NO_x generated from N_2O. Studies of NO_x enhancements following the large Halloween SPEs in October-November 2003 also revealed large quantities of NO_x in the Northern Hemisphere (NH) in January (Jackman et al., 2005). However, dynamical effects, driven by the following major sudden stratospheric warming (SSW)~~in December 2003~~, indicated that this NO_x was unlikely to have originated from the SPEs (Seppälä et al., 2007b). The NO_x increases were more likely a result of the the large amounts of EEP that was present during the polar winter combined with downward descent. For example, Randall et al. (2005) used observations from a number of satellite instruments to show that the springtime NO_x increases in the NH were influenced by strong downward descent in January 2004, bringing excess amount of NO_x down to the stratosphere. Randall et al. (2007) used Atmospheric Chemistry Experiment-Fourier Transform Spectrometer (ACE-FTS) together with HALOE observations to show that the peak upper stratospheric EPP-NO_x was highly correlated with EEP levels over 1992–2005, up until September in the SH. Dynamics influencing the polar vortex is one of the main reason relations between NO_x observations and EPP break down. This is particularly important in the NH, where the polar vortex is more susceptible to SSWs. Randall et al. (2007) further found that the largest EPP-NO_x occurred during the declining phase of the solar cycle, during which more high-speed solar wind streams, driving EEP, are likely to occur.

Using tracer correlations to quantify EPP-NO_x vs NO_x from N_2O oxidation, Randall et al. (2007) suggested that the maximum EPP-NO_x enhancements made up to 40% of the total polar NO_y (total reactive nitrogen, NO_y = NO + NO_2 + NO_3 +

$ClONO_2 + HNO_4 + N_2O_5 + HNO_3$) budget. Seppälä et al. (2007a) contrasted average wintertime A_p levels with the polar winter upper-stratospheric-lower mesospheric NO_x observations from Global Ozone Monitoring by Occultation of Stars (GOMOS) onboard the ~~Envisal~~ Envisat satellite, finding a nearly linear relationship between the two during 2002-2006 for both hemispheres. Funke et al. (2014b) used Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) observations, also from Envisat, of NO_y, to quantify the amount of EPP-NO_y in the polar winter. Analogous to Randall et al. (2007), they found that EPP-NO_y accounted for up to 40% of the wintertime polar NO_y. Funke et al. (2014a) then correlated A_p and EPP-NO_y in the wintertime and concluded that the strong relationship between A_p and EPP-NO_y supports using A_p as a proxy for tracking EPP-NO_y production in the SH wintertime.

1.3 This work

Here, we use stratospheric NO_2 column observations from the Ozone Monitoring Instrument (OMI) on-board the Aura satellite to investigate EPP as a source of NO_x variability in the Antarctic later-winter - spring. ~~Previous work suggested that EPP-NO_x makes up around 40% of the polar winter NO_x.~~ We have a relatively long satellite period (2005-2017) in which to analyse how this NO_x propagates in the following springtime, and whether this is detectable in the NO_2 column. We also analyse how the phase of the QBO affects the contribution of EPP-NO_2 to the total NO_2 column in springtime~~, due to the modulation of the~~. The QBO influence could be likely due to a combination of two effects: 1) QBO phase influences transport of N_2O ~~source.~~ to the polar region (Strahan et al., 2015) resulting in a increased background NO_x source at key times and key altitudes during easterly QBO years (opposite for westerly QBO); 2) QBO conditions influence the probability of PSC formation, and as a result EPP-NO_x may be less/more likely to be removed by denitrification during the winter/early spring during easterly/westerly QBO years. To test for the latter, we will analyse HNO_3 and temperature observations from the Microwave Limb Sounder (MLS), also on-board Aura.

2 Data sets

3 ~~Observations and Methods~~

2.1 OMI NO_2 observations

We use stratospheric NO_2 column observations from the Dutch-Finnish built Ozone Monitoring Instrument (OMI) on-board ~~the~~ NASA's Aura satellite from August to December during years 2005–2017 (v3, Level 2 daily gridded NO_2, see Krotkov (2012); Krotkov et al. (2017)). The daily gridded data has $0.25° \times 0.25°$ horizontal resolution. In our analysis, we use data from latitudes poleward of 50°S. Note that all latitudes henceforth are geographic latitudes. Aura is in a Sun-synchronous orbit in the "A-train" constellation (orbital altitude of 705 km, inclination of 98°, ~~thus~~ 16 day repeat cycle), with ascending node crossing the equator approximately at 1:45pm daily (Schoeberl et al., 2008). As a result, OMI measurements take place at the same locations each year. While NO_2 notably has a diurnal cycle, using observations from the same sunlit locations (thus same local times) each year minimises the effect of this in our analysis.

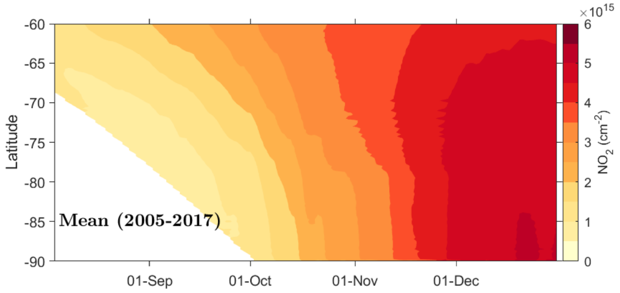

Figure 1. OMI 3 day running mean zonally averaged NO$_2$ column for the time period 2005–2017. The contour interval is 0.5×10^{15} cm^{-2}. The white area at high latitudes in August-September indicates polar night conditions where OMI observations are not available.

The OMI-NO$_2$ data is provided as total column, as well as separated tropospheric and stratospheric columns. This separation is based on the location of the tropopause. Here, we use the OMI stratospheric column observations only. The effective vertical range of the stratospheric column based on the OMI averaging kernels corresponds to $\sim 15-35$ km. At these altitudes, NO$_2$ makes up about 80% of the total NO$_x$ during daytime (see Brasseur and Solomon, 2005, chapter 5.5). As this corresponds to a large fraction of the total NO$_x$, we take the OMI NO$_2$ column measurements to represent a reasonable proxy for the variation in total NO$_x$. The algorithm for the OMI column separation is described in Bucsela et al. (2013). OMI measures back-scattered solar radiation from the atmosphere. Thus, observations are only available for solar illuminated locations – there is no coverage during polar night conditions. ~~This horizontal~~ The latitudinal coverage is illustrated in Figure 1, which presents the zonally averaged mean NO$_2$ column for the period under investigation (2005–2017). ~~This~~ The figure shows how the NO$_2$ column varies in the polar springtime, with increasing amounts of NO$_2$ in the stratosphere as time progresses due to release from its reservoirs (Dirksen et al., 2011). ~~The latitudinal coverage of the measurements is illustrated here, with the lack of measurements during the polar night leading to a gap at the highest latitudes during August-September.~~ The error in the individual NO$_2$ column measurement is estimated to be $< 2 \times 10^{14}$ molecules cm^{-2}, however in areas with low levels of tropospheric pollution (such as the Southern polar region), this error is considerably less (Bucsela et al., 2013). Since June 2007, OMI-NO$_2$ has experienced an issue known as the row anomaly (RA) affecting certain fields of view. All RA affected measurements have been excluded here, leaving around 2×10^5 observations poleward of 60°S per day for the analysis period.

The Aug-Dec monthly mean polar (60°S to 90°S) average zonal mean NO$_2$ columns for each year are listed in Table 1.

2.2 MLS ~~HNO$_3$~~ observations

We use HNO$_3$ ~~profiles from the~~ and temperature profiles from NASA's Microwave Limb Sounder (MLS) which is also on-board the Aura satellite ~~(Manney et al., 2015)~~(Manney et al., 2015; Schwartz et al., 2015). This study uses the version 4.2 product with data screened according to Livesey et al. (2017). ~~MLS HNO$_3$ profiles hve been validated by Santee et al. (2007).~~ The latitude range used here is 60°S to around 82°S and the pressure range used is approximately 100 hPa to 10 hPa. ~~This means~~ MLS HNO$_3$ profiles have been validated by Santee et al. (2007), using data from both the HNO$_3$ 240–GHz radiometer (for

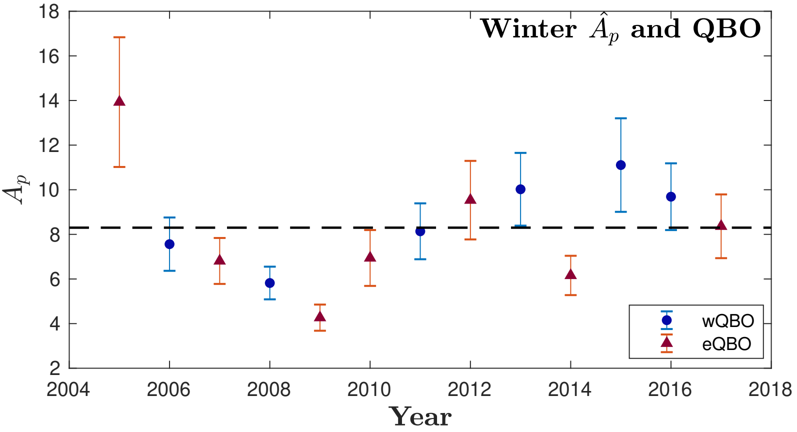

Figure 2. Mean wintertime A$_p$ ($\hat{A}_p$) for each year of this study with error bars indicating 2 times the standard error in the mean. Dashed line indicates the average $\hat{A}_p$ for this study (8.3). The direction of the QBO at 25 hPa in May is indicated with red ~~circles~~ triangles representing eQBO and blue ~~triangles~~ circles wQBO.

pressures ≥ 22 hPa) and HNO$_3$ 190–GHz radiometer (for pressures ≤ 15 hPa). MLS HNO$_3$ has vertical resolution of 3-4 km in the lower – middle stratosphere (used here) and the precision of individual profiles is around 0.6 ppbv in this region. The estimated error in these profiles is no more than 10%. MLS temperature profiles have been validated by Schwartz et al. (2008) with precision of around ±0.6 K on individual profiles between 100 hPa and 10hPa.

2.3 EPP proxy

The geomagnetic activity index A$_p$ is a well-established proxy for EPP ~~(see e.g. Matthes et al., 2017; Funke et al., 2014b)~~ (see e.g. Matthes et al., 2017; Funke et al., 2014a) and is used here to estimate the overall levels of EPP for each polar winter under investigation. During 2005–2017, the mean of winter A$_p$ was ~~8.5~~8.3, reflecting the relatively low overall solar activity during solar cycle 24 (solar cycle 23 average was 12.9). To estimate the overall EPP activity during each winter, we calculate the mean A$_p$ for the period of May–August of each year. These means are hereafter referred to as $\hat{A}_p$ and are provided in Table 1. We designate high $\hat{A}_p$ (H-$\hat{A}_p$) winters as those with ~~$\hat{A}_p > 8.5$~~$\hat{A}_p > 8.3$, i.e. $\hat{A}_p$ higher than the average for 2005–2017. Similarly, we take low $\hat{A}_p$ winters (L-$\hat{A}_p$) as those with ~~$\hat{A}_p < 8.5$~~$\hat{A}_p < 8.3$. The variation in winter $\hat{A}_p$ throughout this study is shown in Figure 2. This figure captures the 11-year solar cycle fairly well, with minimum around 2009 and maximum around 2015.

Several previous works have used tracer correlations to extract solely EPP produced NO$_x$ (or NO$_y$ when information on the NO$_x$ reservoirs is available) (see e.g. Randall et al., 2007; Funke et al., 2014b). When tracer information is not available, other works have investigated the variability in odd nitrogen resulting in variability in EPP levels (usually proxied by A$_p$) (see e.g. Seppälä et al., 2007a). Here, we focus on finding evidence of EPP contribution to column observations. As we do not have mesosphere-stratosphere descent tracer observations available from OMI, we are unable to use the tracer correlation

Table 1. May-Aug mean A_p ($\hat{A}_p$) $\pm 2\times$ standard error in the mean, the polar (60°S to 90°S) Aug-Dec monthly mean, cos($latitude$) weighted, zonal mean stratospheric NO_2 column density ($\times 10^{15}$ cm^{-2}), and QBO phase (E for easterly, W for westerly) for each year 2005-2017.

Year	$\hat{A}_p$	Aug NO_2	Sep NO_2	Oct NO_2	Nov NO_2	Dec NO_2	QBO
2005	13.9 (±2.9)	~~1.12~~ 0.43	~~1.67~~ 0.49	~~3.09~~ 0.84	~~4.71~~ 1.16	~~5.08~~ 1.24	E
2006	7.6 (±1.2)	~~0.96~~ 0.59	~~1.22~~ 0.37	~~2.36~~ 0.69	~~3.54~~ 0.96	~~5.01~~ 1.22	W
2007	6.8 (±1.0)	~~1.09~~ 0.41	~~1.56~~ 0.47	~~3.00~~ 0.82	~~3.80~~ 0.98	~~4.77~~ 1.18	E
2008	5.8 (±0.7)	~~0.95~~ 0.35	~~1.22~~ 0.37	~~2.71~~ 0.77	~~3.79~~ 0.99	~~4.92~~ 1.20	W
2009	4.3 (±0.6)	~~1.08~~ 0.40	~~1.31~~ 0.42	~~2.75~~ 0.77	~~3.81~~ 1.00	~~5.11~~ 1.25	E
2010	6.9 (±1.3)	~~1.13~~ 0.43	~~1.50~~ 0.45	~~2.76~~ 0.78	~~3.80~~ 1.00	~~4.60~~ 1.14	E
2011	8.1 (±1.3)	~~0.98~~ 0.37	~~1.25~~ 0.39	~~2.24~~ 0.66	~~3.78~~ 0.96	~~4.81~~ 1.18	W
2012	9.5 (±1.8)	~~1.23~~ 0.46	~~1.67~~ 0.51	~~3.51~~ 0.90	~~4.45~~ 1.09	~~4.83~~ 1.19	E
2013	10.0 (±1.6)	~~1.04~~ 0.39	~~1.56~~ 0.46	~~3.23~~ 0.87	~~4.74~~ 1.15	~~5.04~~ 1.23	W
2014	6.2 (±0.9)	~~1.03~~ 0.39	~~1.40~~ 0.46	~~2.71~~ 0.76	~~4.17~~ 1.03	~~4.96~~ 1.22	E
2015	11.1 (±2.1)	~~0.94~~ 0.37	~~1.34~~ 0.40	~~2.15~~ 0.63	~~3.69~~ 0.98	~~4.62~~ 1.13	W
2016	9.7 (±1.5)	~~1.10~~ 0.41	~~1.45~~ 0.44	~~2.74~~ 0.75	~~4.39~~ 1.07	~~4.79~~ 1.16	W
2017	8.4 (±1.4)	~~1.08~~ 0.41	~~1.56~~ 0.46	~~2.72~~ 0.74	~~4.51~~ 1.09	~~4.88~~ 1.19	E

methods. To overcome this we perform correlation analysis both for latitudinal coverage and polar average NO_2 observations to find evidence of A_p driven variability in the Antarctic NO_2 column. All correlations between the NO_2 columns and $\hat{A}_p$ are based on the Spearman rank correlation (Spearman ρ), as it more robustly accounts for any non-linear relationships (Wilks, 2011) while still interpreting linear trends where present. Statistical significance is here defined as correlations significant at $\geq$95% ~~.~~ (i.e. p-value of ≤ 0.05).

2.4 Quasi Biennial Oscillation

To investigate the potential QBO effect in the Antarctic atmosphere, we estimate the phase of the QBO from the 25 hPa level zonal mean zonal wind (Naujokat, 1986) near the equator in May each year~~(Strahan et al., 2015)~~. For use of the 25 hPa level in the Southern Hemisphere, see Baldwin and Dunkerton (1998). We ~~take months where the~~ designate years when the zonal mean zonal wind direction is easterly as easterly QBO (eQBO), and westerly as westerly QBO (wQBO). The QBO direction for each year of the study is indicated in both Table 1 and Figure 2. Figure 2 illustrates the approximately biennial nature of the oscillation, with the direction changing almost every year.

3 Results

3.1 NO$_2$ anomalies

~~a) mean NO$_2$ anomaly for years with both H-$\hat{A}_p$ and eQBO. Contour level is 0.2×10^{15} cm^{-2} with black contour representing zero anomaly. b)-d) as b) but different combinations of $\hat{A}_p$ and QBO (see Figure). Figure 3 presents the average anomaly , i.e. the mean deducted~~ We first investigate the anomaly from the mean for each of the four different categories of this study, eQBO H-$\hat{A}_p$, eQBO L-$\hat{A}_p$, wQBO H-$\hat{A}_p$, and wQBO L-$\hat{A}_p$. This is to show how NO$_2$ column evolves in the springtime in the different conditions and to further justify the splitting of years based on QBO phase. Figure 3 presents the average anomaly, (i.e. the mean as in Figure 1 deducted) for each of the four different categories of this study. We can see that winter $\hat{A}_p$ affects the column NO$_2$ present in the spring: years with H-$\hat{A}_p$ (panels a and c) having more positive anomalies from August to November especially in the highest latitudes. In b) and c), the month of October is highly variable with both showing regions of positive and negative anomaly. For low $\hat{A}_p$ years (panels b and d) early spring is not consistently positive or negative, however November displays negative anomalies at high latitudes. The ~~QBO influence~~ combined influence of QBO and $\hat{A}_p$ appears to be most significant for H-$\hat{A}_p$ eQBO years, and wQBO L-$\hat{A}_p$ years (panels a and d). These show consistent but opposite behaviour throughout the spring, with H-$\hat{A}_p$ eQBO years the most favourable for NO$_2$ and wQBO L-$\hat{A}_p$ the least.

3.2 Mean SH polar columns

Figure 4 presents the $\hat{A}_p$ and the mean polar (60°S–90°S) NO$_2$ column for each year (2005-2017) and for each individual month from August to December. ~~The~~ This is to investigate whether there is a relationship $\hat{A}_p$ size and NO$_2$ column, and how this is affected by QBO phase. The mean polar columns are area weighted by cosine of latitude. The phase of the QBO in the preceding May is indicated with red triangles corresponding to eQBO and blue circles to wQBO conditions. ~~Linear~~ Least squares linear fits for all years, wQBO, and eQBO years are included in each panel to guide the eye. The Spearman correlation coefficient (ρ) for each month for all years (yellow) along with eQBO (red) and wQBO (blue) years only are also included in the panels. Note that, as the OMI measurement field gradually increases from an initial maximum latitude of around 68°S in August to 90°S by the end of September, the total NO$_2$ column values do not initially fully encompass the entire polar region (60°S–90°S). The missing data in August and September is treated as missing values in the mean calculation and as such does not contribute to the mean in these figures.

~~As Figure 4, but with monthly average polar NO$_2$ expressed in gigamole (Gmol). Each panel shows the linear least squares fit to data points (colour coding as before), including the fit equations (y =NO$_2$, $x = \hat{A}_p$).~~

The results shown in Figure 4 suggests that a correlation between $\hat{A}_p$ and the stratospheric NO$_2$ column occurs in August, September and November. This is consistent with Figure 3. Furthermore, there is a clear positive correlation for eQBO years from August to November, while for wQBO years, the positive correlation in August and September disappears in October. While the wQBO November linear fit is close to the total fit, the individual years show large variability. In general, wQBO years have consistently lower column NO$_2$ values, especially in August and September. The generally reduced levels of NO$_2$ during wQBO conditions is compatible with the analysis of Strahan et al. (2015), which indicated that the altitudes where

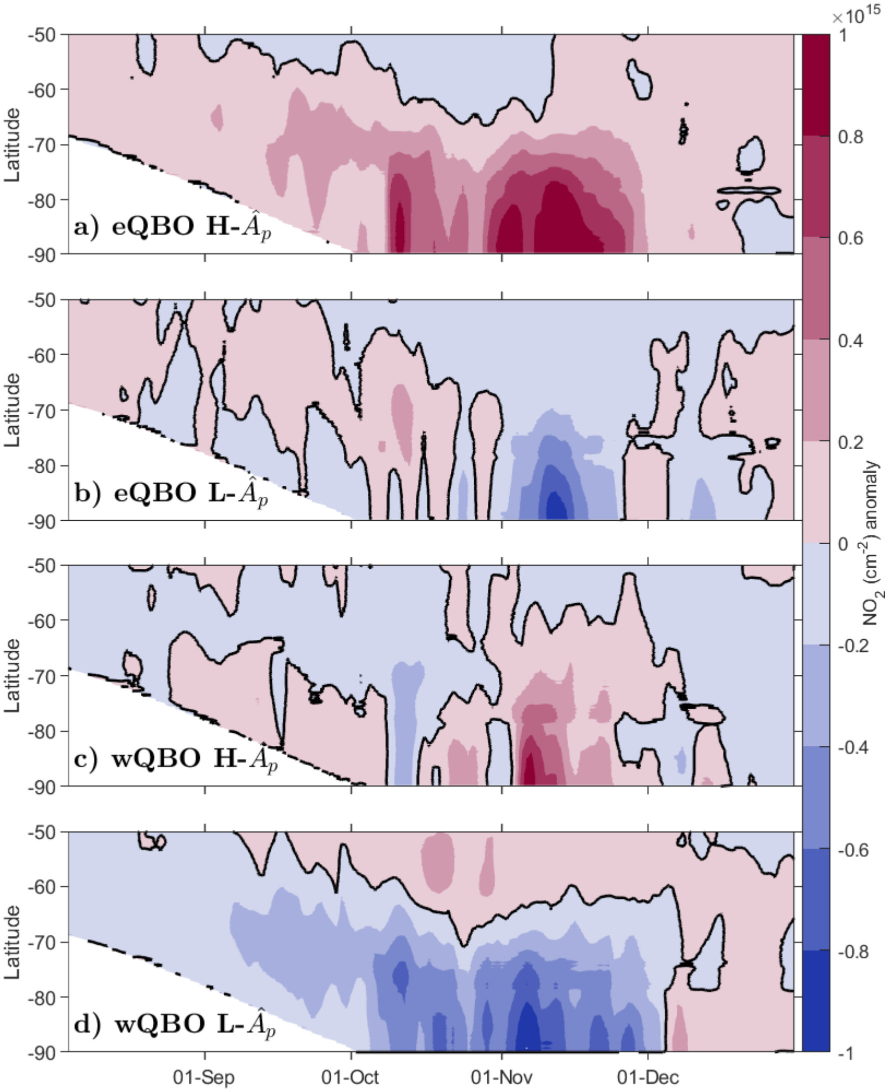

Figure 3. a) mean NO_2 anomaly for years with both H-$\hat{A}_p$ and eQBO. Contour level is 0.2×10^{15} cm^{-2} with black contour representing zero anomaly. b)-d) as b) but different combinations of $\hat{A}_p$ and QBO (see Figure).

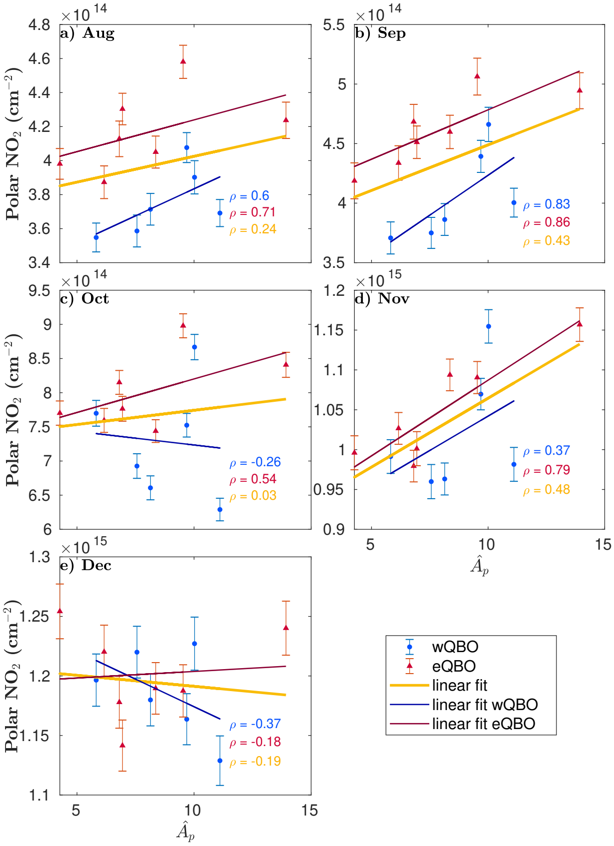

Figure 4. $\hat{A}_p$ versus monthly average, $\cos(latitude)$ area weighed NO$_2$ column density averaged over 60°S to 90°S (where available, see text) for the months of August to December (months as shown in each individual panel). Red triangles indicate years with eQBO and blue circles years with wQBO. ~~Yellow~~ The yellow line shows a least squares linear fit to all data, red line eQBO years only, and blue line wQBO years only. The Spearman ρ (correlation coefficient) for each set is shown in each with corresponding colour, e.g. red ρ corresponds to correlation coefficient for eQBO years etc. ~~.~~ Error bars are 2× the standard error in the mean.

Funke et al. (2014a) reported EPP-NO$_y$ enhancements in later winter-early spring, have consistently lower/higher levels of the dominant NO$_x$ source, N$_2$O, during wQBO/eQBO.

To contrast our results with previous extensive work by Funke et al. (2014a) we ~~now repeat~~ repeated the analysis presented in Figure 4 using the units of gigamole (Gmol, see Funke et al. (2016)) for the monthly mean polar NO$_2$ columns~~.~~). This figure is included in the appendix as Figure A1 ~~again includes~~ and shows the least squares linear fits, ~~now~~ with the corresponding parameters given in each panel. This allows us to estimate the ~~EPP-NO$_x$~~ EPP contribution to the lower stratosphere ($\sim$ 15 – ~~35 km)~~ 35 km) NO$_2$ in the spring, analogous to Funke et al. (2014a). For example, in September (Figure A1, panel b), ~~we see that~~ the approximate contribution to polar stratospheric NO$_2$ column from EPP in eQBO years is ~~+0.021~~ +0.023 Gmol/$\hat{A}_p$. The largest contribution to OMI lower stratospheric NO$_2$ from EPP occurs in November~~for both eQBO and wQBO years as well as all years in general~~, with the corresponding values of ~~+0.058, +0.055 and +0.052~~ +0.073 (eQBO), +0.072 (wQBO) and +0.066 (all years) Gmol/$\hat{A}_p$, respectively. Funke et al. (2014a) (see their Figure 10, showing excess EPP-NO$_y$ in the stratosphere – lower mesosphere in early spring) found an increase in SH polar EPP-NO$_y$ of around +0.0698 Gmol/A$_p$ unit in September. Contrasting this to our results of ~~up to +0.058~~ +0.066 Gmol/$\hat{A}_p$ in November, it seems a large fraction of the EPP-NO$_y$ detected by Funke et al. (2014a) is maintained in the polar region and able to reach the lower stratosphere ~~by November.~~ where it can be detected as NO$_2$ still in November. Note that Funke et al. (2014a) use a weighted A$_p$ scheme.

3.3 Latitudinal correlations

Figure 5 shows the latitudinal extent of the correlation between $\hat{A}_p$ and the 7 day running mean NO$_2$ column for latitudes 50°S–90°S averaged over 1° latitude bins. Stippling indicates correlation significant at $\geq$95% level. This shows how correlation between $\hat{A}_p$ and NO$_2$ column evolves over time and latitude, and different QBO phase. Panel a) presents the correlation when all years are taken into account. Significant positive correlation occurs in late August and variably throughout September, then again in November. October and December show little to no significant correlation. Panel b) shows the correlation for eQBO years only. There are areas of statistically significant positive correlations in all months except December. In October, significant correlations occur only at the very beginning of the month. High positive correlations are still present across 60°S to 90°S from early to mid-November providing first evidence of the indirect EPP-NO$_x$ effect lasting well into the SH spring season. Figure 5 c) presents correlation for years with wQBO only. While positive correlations are present throughout August and September, only small regions are found to be statistically significant. October marks a shift towards negative correlation (not statistically significant) at all latitudes. In November the correlations turn positive once more, but these are again, not statistically significant.

The results ~~from Figure~~shown in Figure 5 suggest that the NO$_2$ ~~column~~ increases at high polar latitudes in September are due to ~~higher than average~~ increased EPP/geomagnetic activity, as strong correlations between NO$_2$ and $\hat{A}_p$ occur in all panels. They also imply that increases in NO$_2$ in November can be due to a combination of high EPP activity and eQBO, whereas wQBO appears to reduce the ~~significance~~ occurence of any EPP induced NO$_2$ increases.

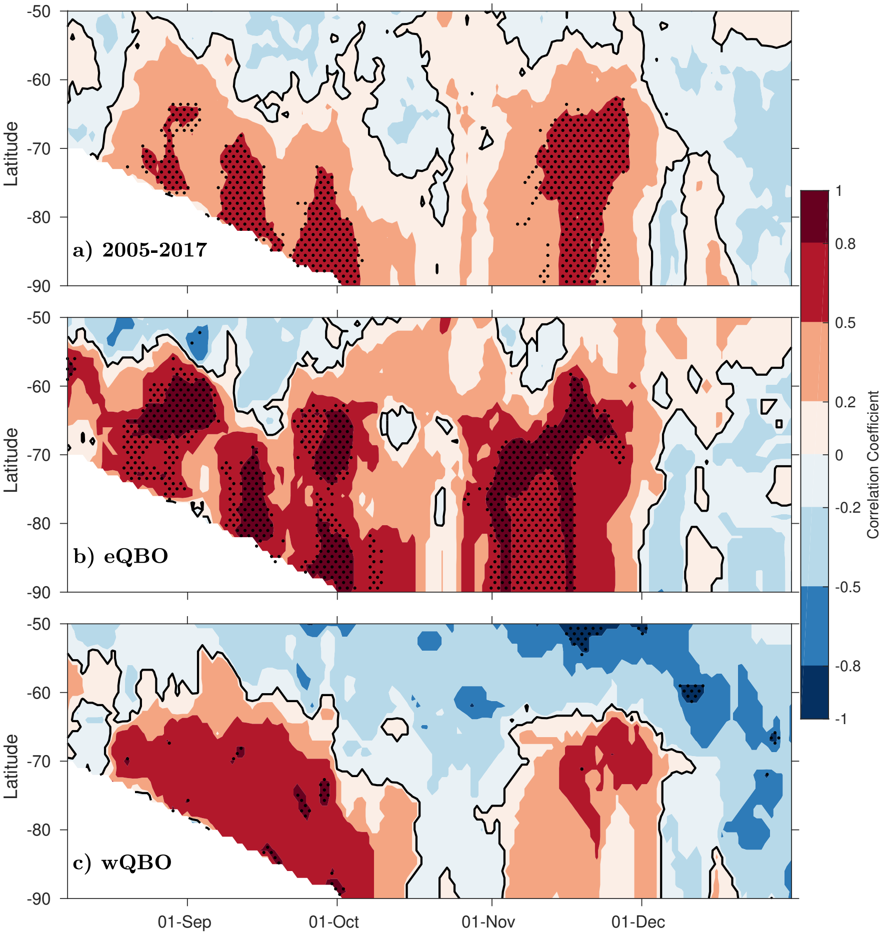

Figure 5. a) Correlation of $\hat{A}_p$ and 7 day running mean NO_2 column density for Aug-Dec for all years. b) Correlation of $\hat{A}_p$ and 7 day running mean NO_2 column density for years with eQBO only. c) as b) except for years with wQBO only. All figures have 1^o latitude resolution. Contour levels are shown for [-1, -0.8, -0.5, -0.2, 0, 0.2, 0.5, 0.8, 1] and the zero contour is indicated with black. Stippling shows regions of correlation significant at $\geq$95% level.

4 ~~Discussion~~

3.1 Polar vortex influence in October

The correlations presented in the previous sections were found to ~~be less significant in October , only to increase again in November.~~ have fewer occurrences of statistical significance in October than the surrounding months. As this time of year marks the typical breakup period of the polar vortex (Hurwitz et al., 2010) and knowing that the descent of EPP produced NO_x is limited to inside the polar vortex (as previously demonstrated for October by Siskind et al. (2000)), we will now investigate ~~the month of~~ October separately, taking the polar vortex into account.

To account for effects from potential asymmetries in the shape of the polar vortex in October in our zonal mean calculations, we repeated the earlier analysis for measurements located inside the vortex. To establish the location of the edge of the polar vortex we utilised the OMI co-located ozone column measurements (Bhartia, 2012): Ozone depleted air is isolated within the vortex until the vortex break up, typically in late November (Kuttippurath and Nair, 2017). Based on this, we take measurement locations poleward of 50°S with corresponding stratospheric ozone column of < 245 DU to be inside the polar vortex. We perform this method for every day of October in the study period to find the daily vortex extent. This is then used to locate NO_2 that is inside the vortex for every day in October over the study period. An example of how the ozone column and the estimated vortex edge are reflected on the NO_2 column measurement for one day of the study (19th October 2014) is shown in Figure 6. Note that we are unable to use this method on months prior to October due to OMI's viewing method and the ozone column not being sufficiently low to detect a clear vortex edge.

Figure 7 a shows the October results (as in ~~Figures~~Figure 4 c~~and 5 b~~) when only observations inside the polar vortex are included. We find that for eQBO, the observations are now much closer to the linear fit than in Figure 4 c), implying that the earlier disappearance of correlation was likely due to variations in the shape of the polar vortex in October.

Similarly for the horizontal distribution of the correlations (Figure 7 b), we now find ~~strong~~ high correlations for eQBO years throughout October. This again implies that the lack of correlation in October is due to the distorted shape of the polar vortex being smeared out by calculation of zonal means, and the effect of EPP on the NO_2 column is significant through October. The reappearance of correlations in eQBO years in November in Figure 5b) is likely a mixing effect, with the break down of the polar vortex around this time leading to vortex air being mixed with extra-vortex air ~~, and thus the net effect on the~~ resulting in the NO_2 ~~column is still observable.~~

distribution not being as skewed as it was when contained in the vortex. Figure 7 c also shows higher correlation with more instances of significance in wQBO years in October than in Figure 5 c though this is more variable than in eQBO years (which is consistent with Figure 5, that wQBO years show lower correlation). Although the A_p index ~~is generally much lower now~~ has generally been lower in the past decade than in the 1991-1996 period investigated by Siskind et al. (2000), considering observations only in the vortex still shows the same, strong linear relationship found in that study. ~~This implies that solar activity of any level generates a proportional response in reactive nitrogen.~~

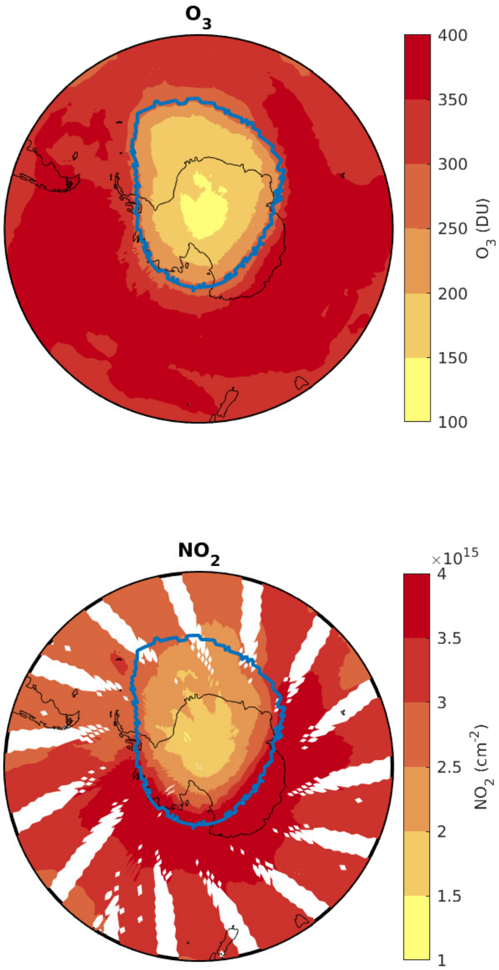

Figure 6. Vortex edge identification based on OMI ozone column ~~.~~ for the 19th October 2014. The top panel shows the OMI measured ozone column density with the 245 DU contour highlighted. The bottom panel shows the NO_2 stratospheric column with the 245 DU ozone contour ~~ovelaid~~overlaid. This method is repeated for each individual day in October throughout the study period.

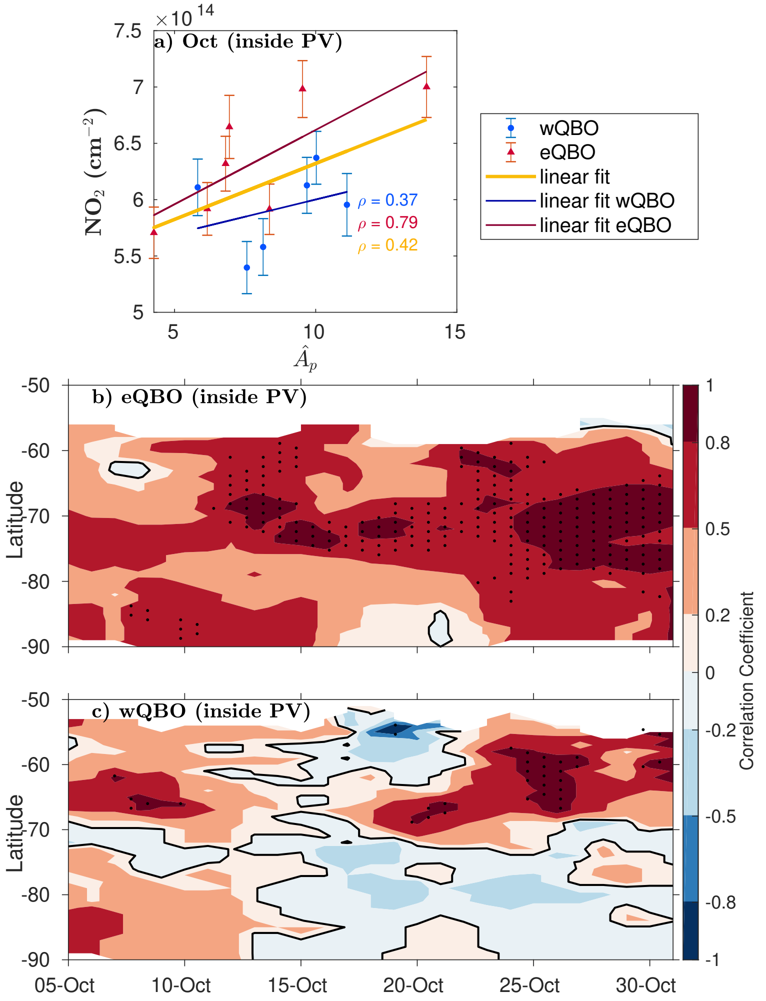

Figure 7. a) $\hat{A}_p$ vs. the average, cos($latitude$) area weighted NO$_2$ from 60°S to 90°S, for observations inside the polar vortex. Red triangles correspond to eQBO years and blue circles to wQBO years. The yellow line represents a linear fit to all data points, while the red line is for eQBO years only and blue for wQBO years only. ρ values are as in Figure 4. Error bars indicate the 95% confidence interval for the mean. b) Correlation of $\hat{A}_p$ with 5 day running mean NO$_2$ column inside the polar vortex for eQBO years. Contour levels are shown for [-1, -0.8, -0.5, -0.2, 0, 0.2, 0.5, 0.8, 1], with an additional black line for the zero contour. The stippling indicates correlations significant at $\geq$95% level. c) as b) but for wQBO years.

4 Discussion

4.1 Influence of the QBO

As shown in Figures 3, 4 and 5 a), our results suggest ~~the that~~ that the phase of the QBO is influencing the SH polar EPP-NO_x signal in the spring months. ~~Strahan et al. (2015) found~~ Results of Strahan et al. (2015) suggest that eQBO phase in early winter leads to ~~decreased~~ increased N_2O ~~in the high polar stratosphere in September.~~ between altitudes of ∼24-33 km in September: Figure 1 of Strahan et al. (2015) ~~clearly shows a negative~~ shows a positive anomaly for N_2O in September in eQBO years ~~for the upper Antarctic stratosphere.~~ (and opposite for wQBO). Although their designation of QBO phase differs slightly from ours, this will not largely affect their comparability, as our designations only differ for one year of a 10 year study. Our results suggest that ~~the lack~~ although the there is increased pool of N_2O ~~transported to the polar regions during eQBO years means that EPP-NO_x contributes more~~ in polar region during eQBO contributing to larger NO_2 column from N_2O oxidation, EPP still contributes a clear fraction to the overall SH polar stratospheric ~~NO_x~~ column NO_2 in the springtime. Taking the QBO phase into account this EPP contribution during spring is more pronounced.

4.2 Possible influence of PSCs and denitrification

~~The affected transport of N_2O does not however explain the obviously depleted in~~ Here we discuss reasons for the consistently lower amounts of NO_2 in wQBO years found in Figure 4~~). This~~. We suggest that this is due to ~~a different effect of the QBO on the~~ the effect of denitrification in the polar region. Baldwin and Dunkerton (1998) found that the polar vortex is colder during winters with wQBO. ~~Colder~~ A colder polar vortex results in a higher likelihood of polar stratospheric cloud (PSC) formation (Brasseur and Solomon, 2005). As discussed ~~earlier~~in the Introduction, PSCs affect the heterogeneous chemistry in the polar region, leading to denitrification of the lower stratosphere (Dirksen et al., 2011). ~~So~~ Thus, for years with more PSCs (i.e. wQBO) ~~we would expect more denitrification to~~ more denitrification would likely occur, resulting in the depleted NO_2 column reported here~~, also explaining~~. This could also explain the lower incidence of significant correlation ~~for these years~~in the wQBO cases.

To test whether QBO ~~direction~~ phase affects denitrification in the Antarctic stratosphere, we ~~use~~ analysed temperature and HNO_3 observations from MLS (see section 2.2). Figure 8 a) ~~shows the mean~~ and b) show the mean temperature and HNO_3 ~~from~~ respectively, each averaged over 60°S to 82°S for 2005-2017 over the late winter–early spring period, i.e. when the polar vortex is coldest and PSCs are forming. Panels ~~b) and c~~c) and e) show the anomalies from the mean temperature, for eQBO and wQBO years respectively. Panels d) and f) present the anomaly from the mean HNO_3 mixing ratio, for eQBO and wQBO years respectively. The vertical pressure range of all ~~three~~ panels is 100 hPa to 10 hPa which corresponds to an altitude range of approximately 17 km to 32 km. ~~As can be seen in~~ Figure 8 ~~,~~ suggest that eQBO years tend to have more HNO_3 (up to 1 ppbv) and higher temperature (up to 4 K) throughout this period, while wQBO years show a consistently negative anomaly ~~(up~~ in HNO_3 (down to −1 ppbv) ~~. This clearly implies that more~~ and lower temperature (down to −4 K). Colder temperatures would likely lead to more PSC formation and thus more HNO_3 ~~is~~ being removed from the stratosphere (more denitrification) in wQBO years (than in eQBO years). It should, however, be noted here that the link of PSC coverage and QBO modulation of

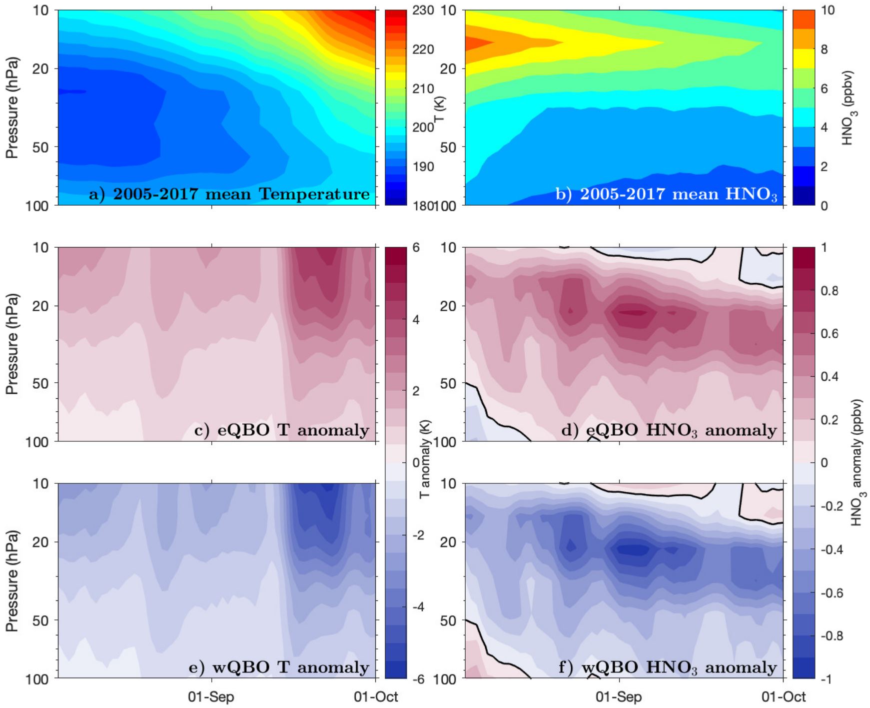

Figure 8. a) ~~3~~ Three day running mean MLS temperature from 2005-2017 (5 K contour interval), b) three day running mean MLS HNO$_3$ ~~volume mixing ratio~~ from 2005-2017 with contour interval 1 ppbv. a) and b) are both averaged over 60°S to 82°S for the lower stratosphere for 2005-2017 late winter–early spring. ~~Contour~~ c) HNO$_3$ anomaly (mean shown in a) has been subtracted) for years with eQBO (contour interval is ~~1~~0.1 ppbv, with black contour showing zero anomaly). ~~b~~d) ~~HNO$_3$~~ Temperature anomaly ~~from the~~ (mean shown in b) has been subtracted) for years with eQBO ~~. Contour~~ (contour interval is ~~0.1~~1 ~~ppbv~~K, with black contour showing zero anomaly). ~~c~~e) as ~~b~~c) but ~~for~~ years with wQBO, and f) as d) for years with wQBO.

polar temperature via the Holton-Tan effects is still under debate (see e.g. Strahan et al., 2015). ~~This supports our hypothesis that wQBO causes more denitrification and thus depletes NO_x, contributing to the results in Figures 3–5.~~

5 Conclusions

We have, for the first time, ~~traced EPP-NO_2 in~~ provided evidence of EPP contribution to the Antarctic stratospheric NO_x column in the late springtime using OMI/Aura stratospheric NO_2 ~~column observations.~~ observations. This is one of the few studies to use stratospheric NO_2 data from OMI and also highlights the value of long time series of stratospheric NO_2 from nadir viewing instruments. Our analysis shows that influence from the QBO is able to mask the stratospheric EPP-NO_x signal in satellite observations in a way that to our knowledge has not previously been accounted for: ~~Increased EPP during the winter, when combined with eQBO phase , results in~~ Accounting for the phase of the QBO makes the contribution from EPP more pronounced in the NO_2 column, and signals of enhanced EPP-NO_x in the polar stratospheric column can be detected until late November.

~~The QBO influence is likely due to a combination of two effects: 1) eQBO reduces transport of N_2O to the polar region (Strahan et al., 2015) resulting in a reduced background NO_x source in eQBO years. Thus enhancements in EPP-NO_x become more evident in the springtime; 2) eQBO conditions reduce the probability of PSC formation, and as a result EPP-NO_x is less likely to be removed by denitrification during the winter/early spring. This is also supported by MLS HNO_3 observations, showing less denitrification during eQBO years.~~

Previously, Funke et al. (2014a) analysed EPP-NO_y observations and were able to attribute SH enhancements with an average A_p dependence of +0.0698 Gmol/A_p into early spring months (September). Here, we show that this reactive nitrogen lingers, entering the lower stratosphere in the form of NO_2 at an average ~~rate of +0.052~~ A_p dependence of +0.066 Gmol/$\hat{A}_p$ in November.

We present evidence of contribution from EPP-NO_x in the Antarctic stratosphere at a time when ~~the ozone hole is present~~halogen activate ozone loss is taking place. NO_x is well known to react with both ozone and active halogens, catalytically destroying the former, and driving the latter to its reservoirs (Brasseur and Solomon, 2005). Antarctic ozone loss has been found to be reduced in years with eQBO (Garcia and Solomon, 1987) due (at least in part) to the increased vortex temperatures hampering chlorine activation on PSCs (Lait et al., 1989). Our results suggest that, as chlorine activation continues to decrease in the Antarctic stratosphere following the Montreal Protocol (Solomon et al., 2016), the total EPP-NO_x (in addition to SPEs as pointed out by Stone et al., 2018) ~~in eQBO years needs to~~ should be accounted for in predictions of Antarctic springtime ozone recovery. Future studies should investigate the ~~effects of~~ role of larger EPP-NO_x fraction when investigating the net effect of NO_2 on the fragile ozone chemistry in the springtime~~and how this is modulated by the QBO~~.

Data availability. All data used here are open access and available from the following sources: A_p: http://wdc.kugi.kyoto-u.ac.jp/kp; QBO: https://www.geo.fu-berlin.de/en/met/ag/strat/produkte/qbo; OMI and MLS: https://earthdata.nasa.gov.

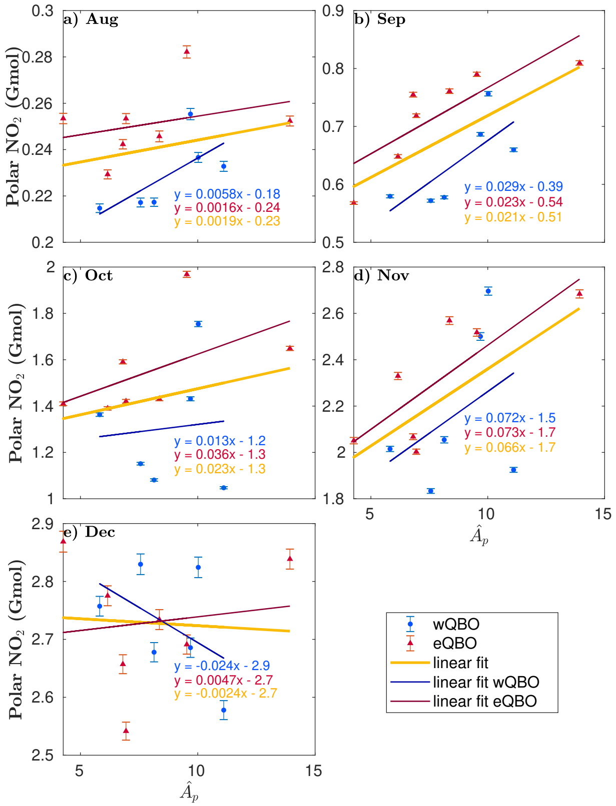

Figure A1. As Figure 4, but with monthly average polar NO$_2$ expressed in gigamole (Gmol). Each panel shows the linear least squares fit to data points (colour coding as before), including the fit equations ($y =$NO$_2$, $x = \hat{A}_p$). Red triangles are years with eQBO, and blue circles are wQBO. The yellow linear fit is a best-fit line for all the data in each plot, while the red fits only eQBO data, and the blue line fits only wQBO data.

Author contributions. EMG and AS planned the study. EMG did the analysis with support from AS. JT provided expertise on OMI observations. EMG and AS lead the writing of the manuscript with comments from all authors.

Competing interests. The authors declare no competing interests.

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