

Author responses to reviewer comments on paper #acp-2019-1035 "EPP-NO_x 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-NO_x 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 NO_x 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 NO_x concentration (produced by N₂O oxidation) and not the EPP contribution. (ii) would represent a total NO_y loss mechanism (independently whether produced by EPP or N₂O) and hence would not alter the relative EPP-NO_x 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-NO_x 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, NO_y 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 am 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 increased 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 HNO₃ 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

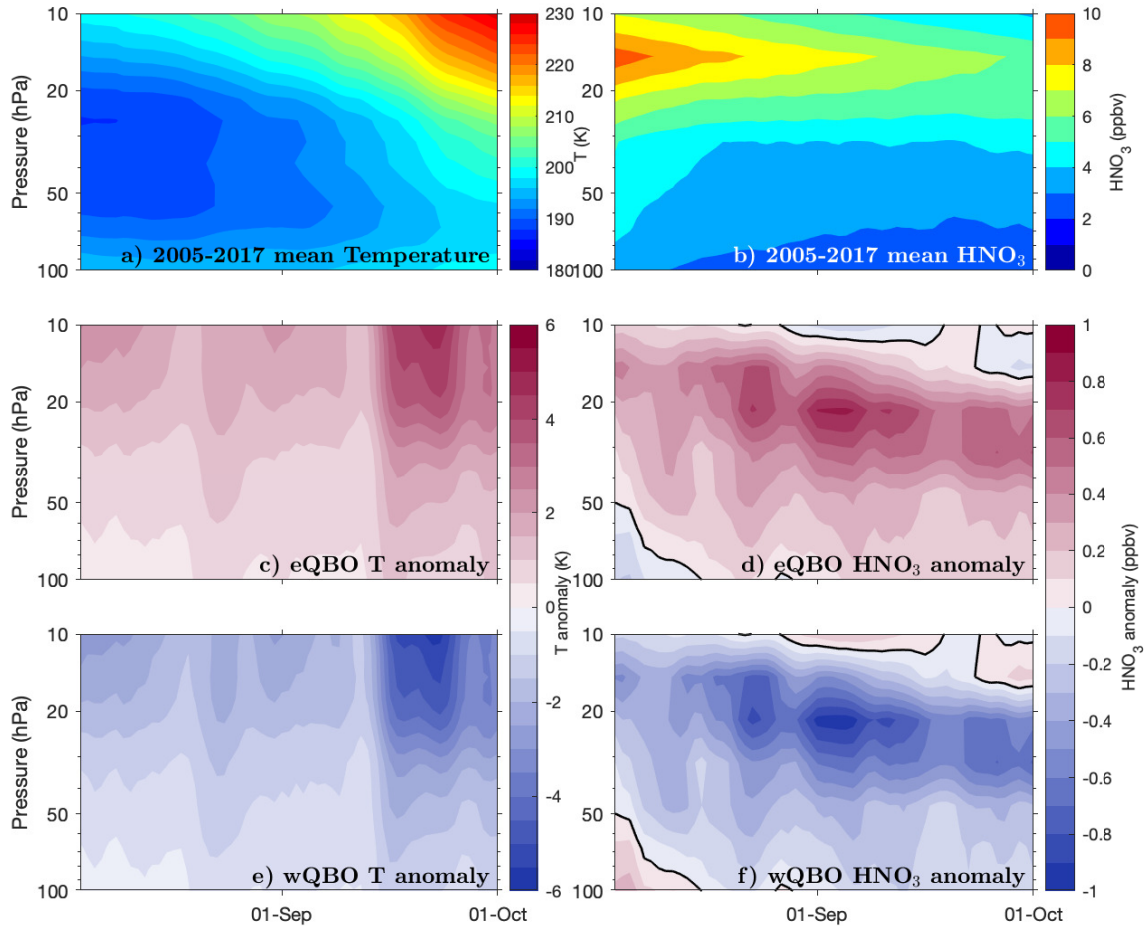


Figure 1: Temperature (left) and HNO_3 (right) mean fields (a-b) for the study period and anomalies for eQBO (c-d) and wQBO (e-f) phases

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

HNO₃ (down to -1 ppbv) and lower temperature (down to -4 K). Colder temperatures would likely lead to more PSC formation and thus more HNO₃ 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: 123-25: 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 N₂O 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 N₂O 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: 127: strictly speaking it is HNO₃ (not NO_x) 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 HNO₃ reservoir

Comment: 163: 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: 185-86: This sentence is a repetition of what is stated in the preceding paragraph.

Reply: We have removed this as suggested.

Comment: 187 "...whether this IS detectable..."

Reply: We have corrected this as suggested.

Comment: 1147: 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: 1186: Figure 5 shows correlations, not NO₂ column increases.

Reply: We have revised this sentence to: *The results from Figure 5 suggest that the NO₂ increases at high polar latitudes in September are due to increased EPP/geomagnetic activity, as strong correlations between NO₂ and \hat{A}_p occur in all panels.*

Comment: 1204-205: 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 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-NO_x 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.

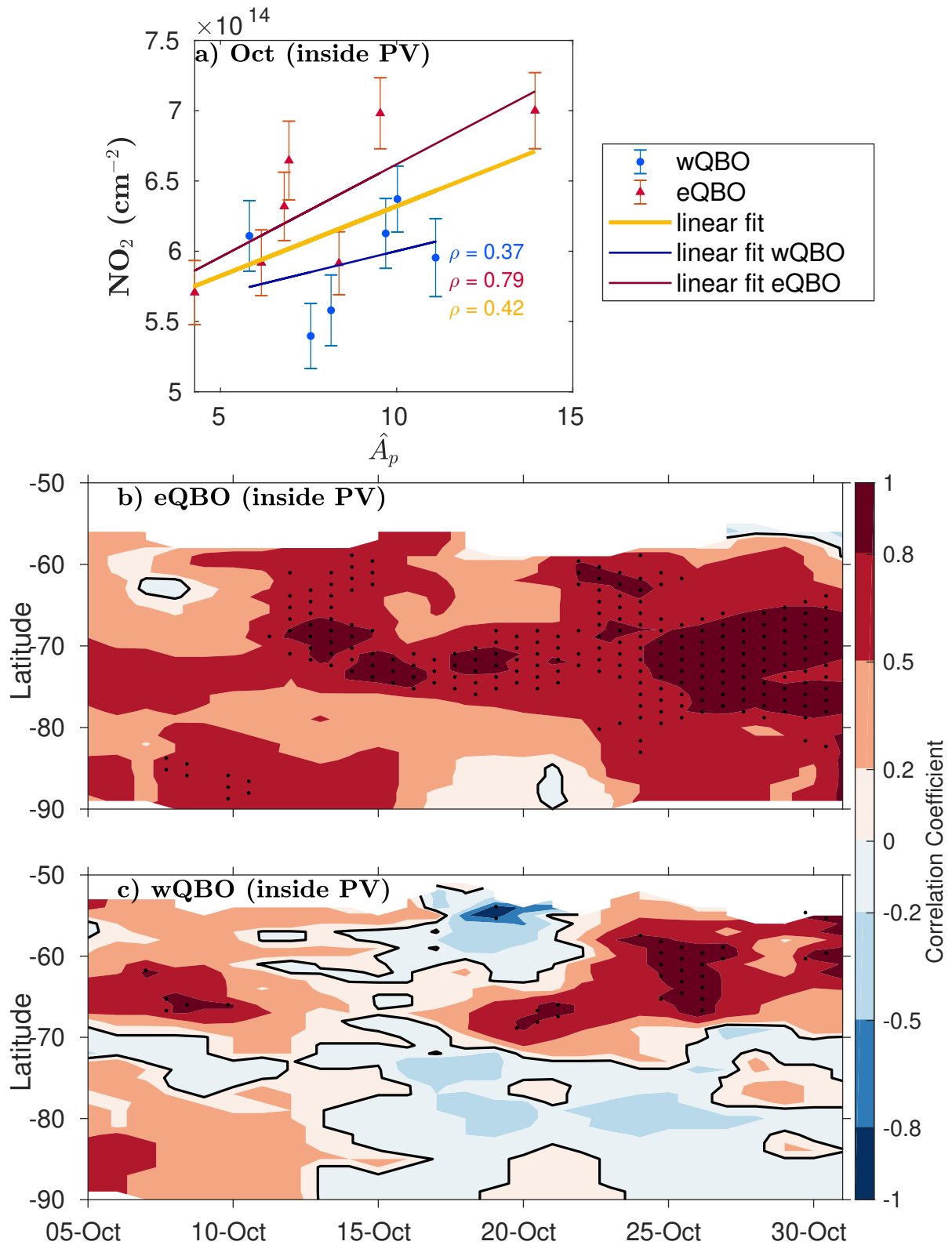


Figure 2: Revised Figure 7, a) now area weighted, b) as before, c) showing the results inside the vortex for wQBO.