

We thank the two anonymous reviewers for their thoughtful comments. To address the reviewers' main concerns, we have revised the manuscript significantly in two aspects:

- 1) We agree with both reviewers that the use of GEOS-5 model CO is distracting and adds nothing to the weight of the main conclusions. We follow the referee #2's suggestion and delete the discussion of GEOS-5 model CO in this study. The manuscript is re-organized with section 3 for the air mass identification and composition, sections 4 and 5 for analysis of NO_y , O_3 , O_3 production within individual air masses sampled during ARCTAS.
- 2) We re-organize section 4 to add a sub-section 4.2 that includes a detailed discussion on $\text{C}_2\text{H}_6 \rightarrow \text{CH}_3\text{CHO} \rightarrow \text{PAN}$ as the main source of PAN in the STE air masses. We show that there are elevated levels of CH_3CHO and active conversion of NO_x and HNO_3 to PAN in two fresh and one aged STE plumes sampled by DC-8 on July 9 and 10 2008 (newly added figures 7 and 8). CH_3CHO can be formed from photochemical degradation of $>\text{C}1$ alkanes (e.g. ethane, propane, n-Butane) and $>\text{C}2$ alkenes (e.g. propene), isoprene, and ethanol. Unlike alkenes, isoprene, and methanol whose contribution to acetaldehyde production are not affected by NO_x , the molar yields of CH_3CHO for alkane family are higher at high NO_x . Among all alkanes, ethane is the mostly source precursor of CH_3CHO since it has longer lifetime. When air of stratospheric origin mixes into the troposphere during STE, it provides high NO_x which stimulates both the production of CH_3CHO from ethane and the production of PAN from CH_3CHO , therefore displays elevated levels of PAN.

Specific responses to additional minor comments:

Anonymous Referee #1

Finally, the Conclusions include comparisons to TOPSE and ABLE-3 measurements; I think this discussion could warrant its own section prior to the Conclusions. Otherwise, I think it would fit better in Section 4 where the ARCTAS average concentrations are discussed. It seems like new material suddenly appearing in the Conclusions in the current version.

We follow the reviewer's suggestion and moved this discussion to Section 4.

Section 3. The under-estimation of the modeled CO is attributed primarily to OH values being too high. Why are not an underestimation of sources considered as a possible cause? Also, the factors accounting for secondary production from HCs could be too low.

The use of GEOS-5 model CO is deleted from this study as suggested by the reviewers (see above).

Fig 3 & 4. It would be nice to have more of a justification given for uniformly adding +25 ppb to the model CO. Is this adjustment used only for visually comparing the distributions, or are the modified values used elsewhere?

The use of GEOS-5 model CO is deleted from this study as suggested by the reviewers (see above).

p.10731, l.5-22: Give more justification for looking at C₂H₂/CO, and/or at the end of the paragraph explain how this ratio will be used later in the paper. I think including the Supplement figures here instead of Fig 5 would be more appropriate. Does C₂H₂/CO really show you something more than CO in Fig. 6? If so, make that clearer. If not, it could be left out.

We agree with the reviewer that the addition of C₂H₂/CO ratio do not add much to that can be shown by CO. We have excluded the discussion of the C₂H₂/CO ratio from this study and delete the original Figure 5 and Figure 6 panels bd (Figure 6 now becomes Figure3)

p.10731, l.20: I think the long lifetime of CO in the winter Arctic is equally, or more, the cause of the well-mixed troposphere than slow transport times.

This paragraph is deleted in the revised manuscript.

p.10737, l.3: You state "we calculate the extent of mixing : : :". Say more about how that calculation is done and how you reach the conclusions in the following sentence.

We have added a figure (Figure 6) using CO as a proxy for transport and air mass inter-mixing, to illustrate how mixing ratios of reactive nitrogen species change as air of stratospheric-origin mixes with tropospheric background during STE, following Parrish et al. (1998). As it mixes with tropospheric air, an air parcel of stratospheric origin moves along the mixing line (thick green dashed lines) in a scatter plot. When active chemical production and/loss of reaction nitrogen species occurs, the air parcel deviates from the mixing line. The majority of the observed mixing ratios of NO_x, HNO₃, NO_y, and the springtime PAN in the STE air masses are within the envelope of variations that can be explained by mixing. Levels of PAN in the summertime STE air are significantly higher than that can be explained by pure mixing with the tropospheric background, indicating active photochemical production. This discussion is added in section 4.2.

Table 2: for STE, do you mean 80<CO<160 and 50<CO<120 ? (missing first "<")

Corrected.

p. 10740, below (R8) in 2 places, should be "production and loss rates" (not "product"). A number of minor grammar errors (singular/plural, missing articles, etc.) that I have not bothered to note.

Corrected.

Anonymous Referee #2

Page 10737 - Discussion of calculations of extent of mixing between stratospheric air and background based on tracer relationships. Please add a brief outline of what was done to determine that the change in NO_y partitioning can be given by mixing only.

We have added a figure (Figure 6) using CO as a proxy for transport and air mass inter-mixing, to illustrate how mixing ratios of reactive nitrogen species change as air of stratospheric-origin mixes with tropospheric background during STE, following Parrish et al. (1998). As it mixes with tropospheric air, an air parcel of stratospheric origin moves along the mixing line (thick green dashed lines) in a scatter plot. When active chemical production and/loss of reaction nitrogen species occurs, the air parcel deviates from the mixing line. The majority of the observed mixing ratios of NO_x, HNO₃, NO_y, and the springtime PAN in the STE air masses are within the envelope of variations that can be explained by mixing. Levels of PAN in the summertime STE air are significantly higher than that can be explained by pure mixing with the tropospheric background, indicating active photochemical production. This discussion is added in section 4.2.

Page 10740 - Box model analysis, Equation R9. Please briefly provide more information to the reader on which terms in these model calculations are constrained and with what.

We add a brief description of the box model calculation:

“We use the O₃ production and loss rates calculated by the NASA Langley box model (Olson et al., 2004) constrained by chemical and physical parameters measured by the DC-8 aircraft. Observed O₃, CO, NO, temperature, J(NO₂) and J(O₃) from the 60-sec merge are used as model input. In addition, model calculations have been constrained by observed values of many trace gases, including H₂O₂, CH₃OOH, HNO₃, PAN, acetone, MEK, methanol, and ethanol when possible. High-resolution (1-sec) simulations are employed when it is determined that high heterogeneity of NO within a one-minute average results in erroneous calculations of HO_x and related radical species. For these points, the one-minute average has been broken into 60 1-sec points for modeling purposes. 1-sec data for NO, O₃, CO, and H₂O are used for these calculations. The high resolution (1-sec) results are then averaged back to the 60-sec merge.”

Page 10741 - P(O₃) rates in STE air masses. It would be interesting to know how the balance between O(¹D) + H₂O loss and HO₂ + NO production is balanced in these air masses as they mix with moist tropospheric air. Can the relevant terms be plotted from Equation R9?

To address this, we have added a figure (Figure 11) and a detailed discussion to illustrate how the production and loss terms ($K_2[HO_2][NO]$, $K_6[H_2O][O(^1D)]$, and $K_7[HO_2][O_3] + K_8[OH][O_3]$) in Equation R9 differ in three types of air mass: Background, Stratospheric air, and STE air.

Abstract, line 4: "the GEOS-5 CO simulation" change to "a GEOS-5 global model CO simulation"

This phrase is deleted.

Abstract, line 9: "with CO decreases" change to "with CO decreasing"

Done.

Page 10728, line 27: "well captures" change to "captures well"

This paragraph is deleted.

Page 10730, line 24: "define" change to "defined"

This paragraph is deleted.

Page 10740, paragraph 1: Change two instances of "product" to "production".

Done.

Table 1 caption: Correct text "used innthis study"

Changed to "used in this study".

Table 2, footnote 'b'. Change "scattering" to "scatter"

Done.