The paper presents two years of NOx, NOy, and PAN data taken at Summit, Greenland along with showing ozone and hydrocarbon data over the same period. This is combined with FLEXPART analysis to understand the influence of anthropogenic and biomass burning (BB) sources. The paper contains very interesting data that should be published, however there are some problems with the manuscript in its current form. The first reviewer has done a very good job pointing out both scientific and technical issues with the manuscript. Therefore, those comments will not be repeated here. Unlike the first reviewer, I find it may be worth to include the NMHC measurements in the paper. This is specifically addressed below (comment 3).

We would like to thank reviewer 2 for the valuable comments. We have addressed each comment separately below.

1. The paper begins by showing two years of data as monthly averages (months 1-12) for NOx, NOy, PAN, and ozone. However, later the entire NMHC record is shown for the same time period later. The authors should decide if they want to focus on averaging together these two years of data as representative of the seasonal cycle (as is done in Figure 1 and 2) or if they want to show the actual time series as the basis for the analysis (Figure 4). In my opinion averaging the two years removes some of the valuable information in this dataset. Why not show NOx, NOy, PAN, and ozone as monthly averages separately for each year in Figures 1 and 2. The same applies to Tables 1 and 2; are the values different for the two years of data, or is the average representative of both years?

The reviewer has made an excellent suggestion. Figure 1 (as shown on the next page) has now been revised to show the data as monthly averages over each year separately. Tables 1 and 2 will also be updated to include the statistics for each year as separate columns. We feel that the updated plots and tables will provide more information on the differences between the two winters which is particularly important for the discussion in section 3.2.1.

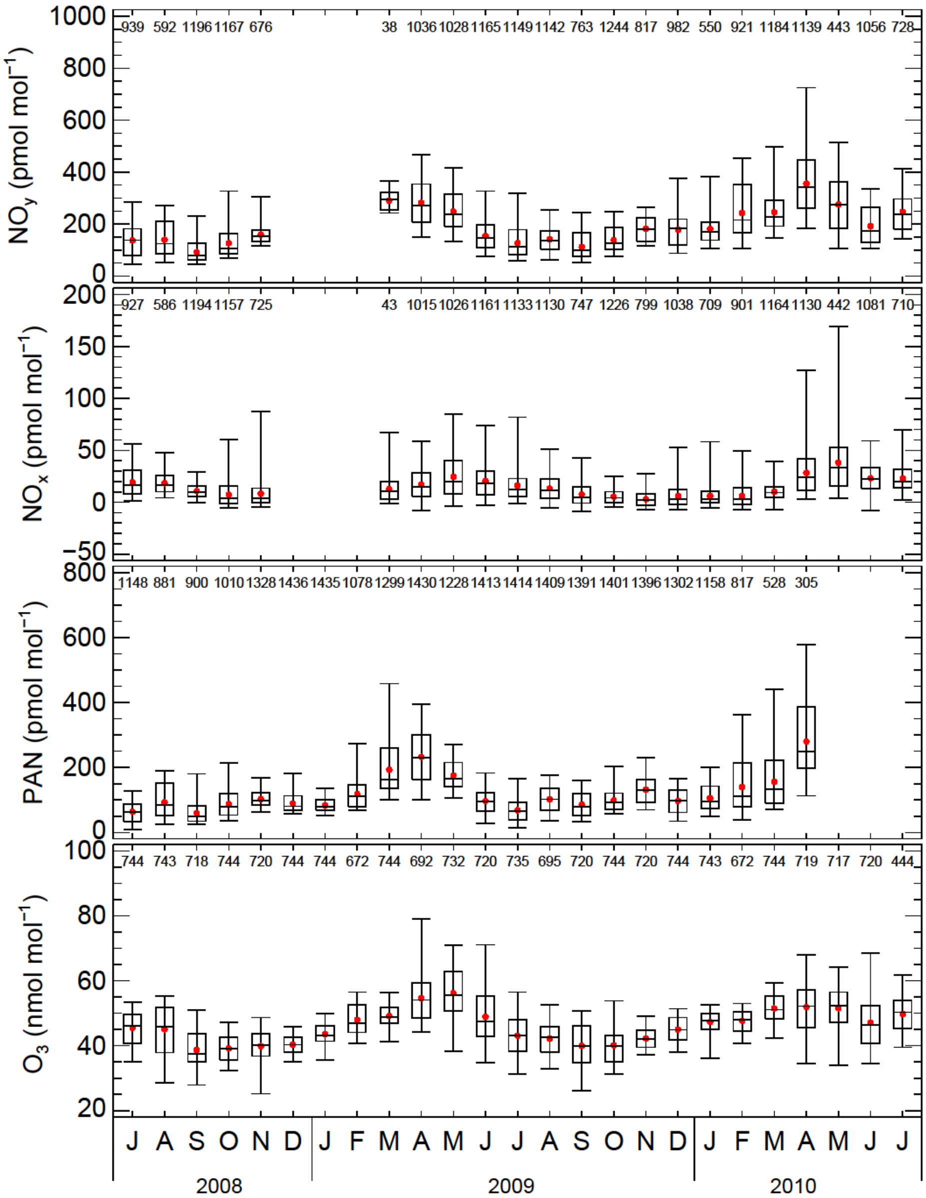

Figure 1. Monthly averages of (a) NO_y, (b) NO_x, (c) PAN and (d) O_3 at Summit from July 2008-July 2010.The median and mean are indicated by a filled red circle and black square, respectively; the box indicates the middle 67% of the data; and the top and bottom of the vertical whiskers indicate the 1st and 99th percentile of all the data. The numbers at the top of each plot represent the number of 30 min averages included in the distribution.

2. Figure 3 is not very useful as it is presented. If the authors want to show some information about the diurnal cycles of NOx, NOy, and PAN they should show data for each month (not a three month average). The amount of sunlight at Summit changes radically between April and June. Given the influence of snow on NOx levels it's not fair to average all of this data together to give one diurnal profile.

We agree with the reviewer and have separated the diurnal plots in Figure 3 to show data for each month. We have also included the diurnal cycle for March to cover the early spring period. The text in the manuscript will be updated to reflect the new plot.

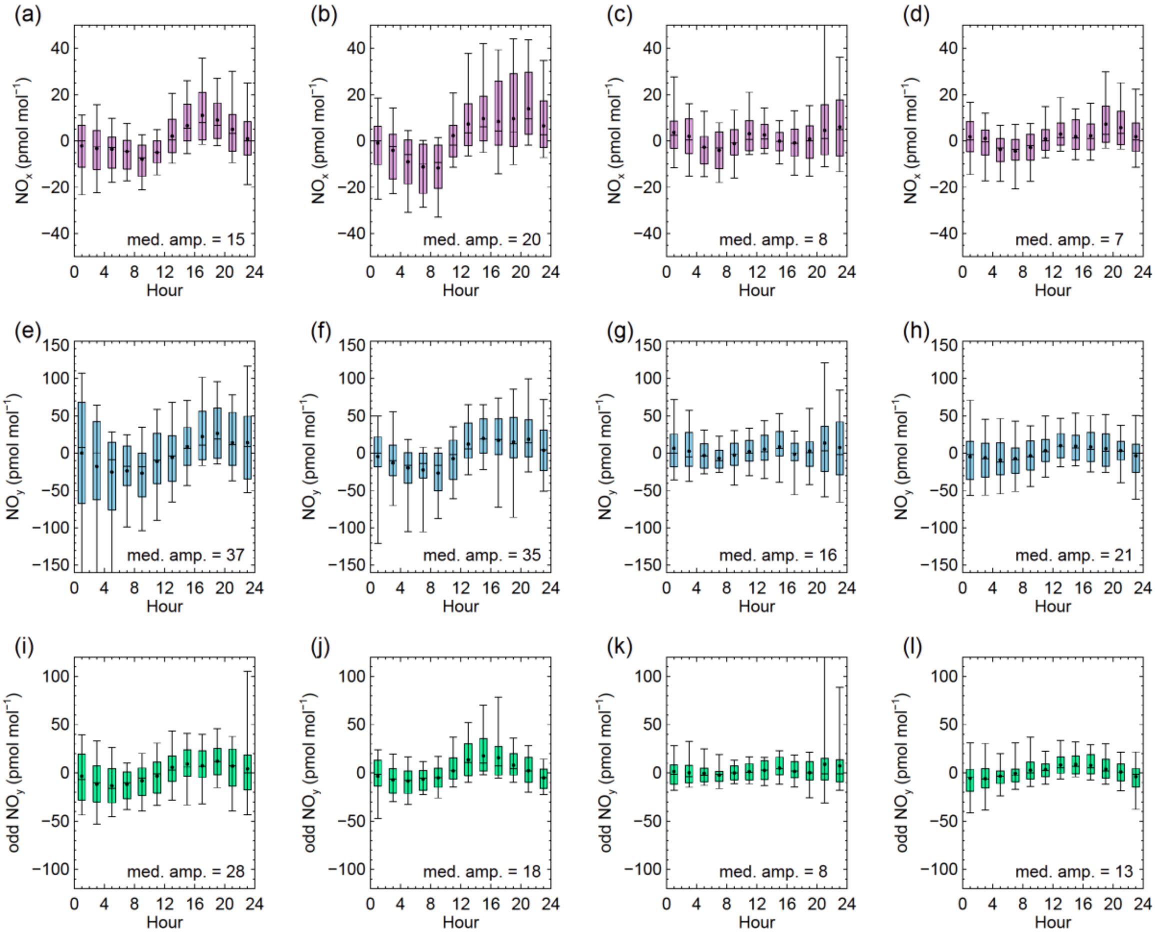

Fig. 3. Average diurnal cycles of ambient NO_x (a,b,c,d), NO_y (e,f,g,h) and odd NO_y (I,j,k,l) measured at Summit for April (1st column), May (2nd column), June (3rd column), and July (4th column). Median ambient levels observed each day have been subtracted, to remove any impact from day to day variability. The median and mean of the data are represented by a filled black circle and horizontal, respectively; the colored box indicates the middle 67% of the data; and the vertical whiskers indicate the middle 95% of the data. Times are shown as UTC-2 hours. The median amplitude of the diurnal cycle (units=pmol mol^{-1}) is noted on each subplot.

3. I do find it appropriate to have some information about NMHCs directly in the paper (even if it's already been published elsewhere). However, the authors should use the same box/whisker plot analysis for Figure 4 as in Figure 1 (two years of monthly average data, with the two years of data presented separately). Otherwise, the plot is almost the same as already presented in Helmig et al., 2014a and provides nothing in addition to what has already been published.

The authors agree with the reviewer and have changed Figure 4 to show the 6 NMHC as box-and–whisker plots separated for the 2 years of measurements, to be consistent with Figure 1. The updated plot is shown in the document containing the responses to reviewer 1.

4. The FLEXPART analysis given in Figures 5 and 6 is questionable given that the paper does not focus on black carbon or aerosols. CO source contributions are a more appropriate choice because they do not suffer from the same wet removal issues.

The BC tracer was used here for the retroplume analysis as simulations are available for both the anthropogenic and fire tracers as part of the POLARCAT campaign. Simulations using CO were only performed for the anthropogenic tracer. Using the BC tracer allowed for consistency between biomass burning and anthropogenic investigations. A comparison was made between the CO and BC anthropogenic tracers to determine whether pollutions events were missed. A time series for the two tracers from summer 2008 to 2010 is shown in Figure R-1 below. Figure R-2 shows a correlation plot of the two tracers, with a Pearson's correlation coefficient, r=0.973. The result suggests that although there may be some differences in the magnitude of the FLEXPART tracers, the transport simulated with the BC tracer is very similar to the CO tracer. The plots below will be included in the supplementary material and a discussion on the comparison will be provided in the revised manuscript.

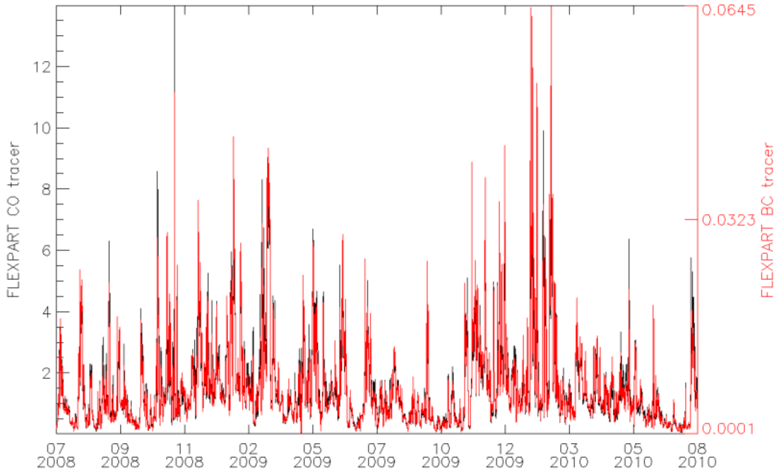

Fig R-1. Time series of the FLEXPART CO tracer (black) and the FLEXPART BC tracer (red) at Summit for July 2008 – July 2010.

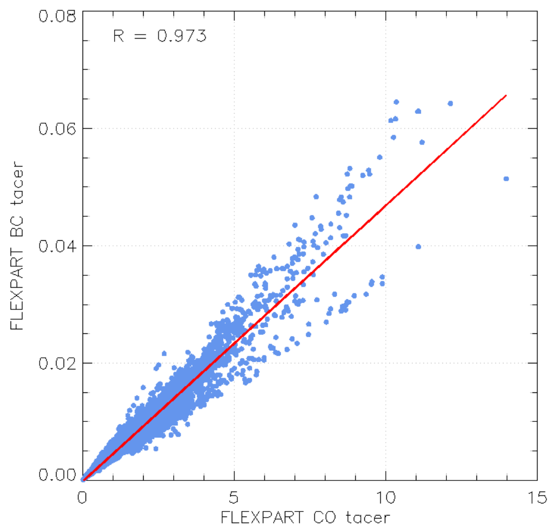

Fig R-2. Correlation of FLEXPART CO tracer and the FLEXPART BC tracer at Summit for July 2008 – July 2010. The Pearson's correlation coefficient (R) is shown in the upper left corner of the plot.

5. Figures 4 and 5 also show that anthro and BB sources contribute different amounts during the different years studied. This is further motivation to present the data from each year separately in Figures 1 and 2 (discussed in comment 1). Why are source contributions only shown for December through March? It would be useful to also show a period in summer for the BB tracer contribution.

This is a very good point and the revised manuscript will include a plot showing the monthly source contributions from FLEXPART for each month separately, from 2008-2010. This plot is presented in the document containing the responses to reviewer 1. Additionally, section 3.2 will be re-written to include a detailed discussion on the contributions to the BC_{anthro} and BC_{fire} tracer from different source regions during different times of the year.

6. The paper should specify what version of FLEXPART and what emissions are used for the anthro and BB sources.

We thank the reviewer for noting the missing information regarding the FLEXPART model. This information will be provided in the revised manuscript.

7. Figures 7 and 8 are interesting because they contain a portion of the measured time series, so pollution events can been seen in the data. The authors should include a full time series in the electronic supplement and only show the portion of the time series discussed in detail in the main portion of the paper.

We agree with the reviewer and the full time series of the measurement species and FLEXPART contributions will be presented in the electronic supplement with individual events discussed in the main text identified. A table with details on each anthropogenic event will also be provided in the supplementary material.

8. Figures 9, 10, and 11 use column integrated FLEXPART retroplume analysis to study airmass origin. This is a good approach to understand features that originate from long-range transport in the NOx, NOy, and PAN data. However, the figures as presented are confusing. What information does the altitude of the plume provide (given that the gray points are hardly visible on the plots)? Is the gray the altitude 10 days prior to release, or the altitude where the particles reside for more than ten days? This portion of the paper is very confusing.

The figures of the FLEXPART retroplume include 10-day particle centroid locations. Only the locations from 1-10 days upwind are shown on the plot for simplification, however, retroplumes include simulations 20 days prior to arriving at Summit. FLEXPART is not a back-trajectory model, but rather a Lagrangian Particle Dispersion Model. As such, at any point in time there are thousands (in this case 10s of thousands) of particles in the model domain. The circle also provides an indication of the altitude of the plume at the centroid location of the particles in the model domain N days back from the measurement date, where N is the number shown next to the shaded circles. The shading represents the altitude of the centroid location. A darker circle indicates a lower altitude as given by the gray-scale marker.

The revised manuscript will include this description in the figure captions and the figures will be made clearer for the reader.

9. Figures 10 and 11 study specific events, however the portion corresponding to the particle release is not indicated on the measurement plots. I found myself trying to shade in the periods from 7/26/2008-7/27/2008 and 8/4/2008-8/5/2008 on Figures 10 and 11 the correspond to the FLEXPART release times. The authors should put the effort into making these plots understandable for the reader.

We thank the reviewer for noting this issue with Figures 10 and 11. The plots will be updated and the period corresponding to the release time for the FLEXPART retroplume will be indicated.

10. Page 13838 - Lines 20 to end of page: The case of the FLEXPART BC not coinciding with enhanced PAN, NOy, and ethane is a bit of a mystery. This will be less confusing if the authors change to using CO source contributions, since BC is subject to wet removal. CO is a more straightforward to compare with PAN and ethane since they experience more similar atmospheric processing. If the disagreement still persists after looking into CO by source, then the authors should look into differences in plume altitude compared with other similar plumes. Are there emissions missing that can explain this? Is there a difference in transport pathways (e.g. residence time in the boundary layer or upper troposphere/lower stratosphere) that can explain this?

As discussed in the response to comment 4 above, the simulations do not appear to be missing any events when using the BC tracer instead of the CO tracer. Figure R-3 shows a time series of the BC and CO tracer, zoomed in on the period in 2010 that was discussed in the text. There is no indication of additional anthropogenic events in the CO tracer that are not present in the BC tracer. It is possible that FLEXPART is missing some sources, or, as mentioned in the manuscript, the pollution originated prior to the 20 day simulation. The retroplumes for these periods have been investigated, however, a comparison between these events and those when FLEXPART agrees with the data have not been performed so far. We thank the reviewer for providing an excellent suggestion and a comparison of plume altitudes and transport pathways for the different events will be performed and the manuscript updated with the results.

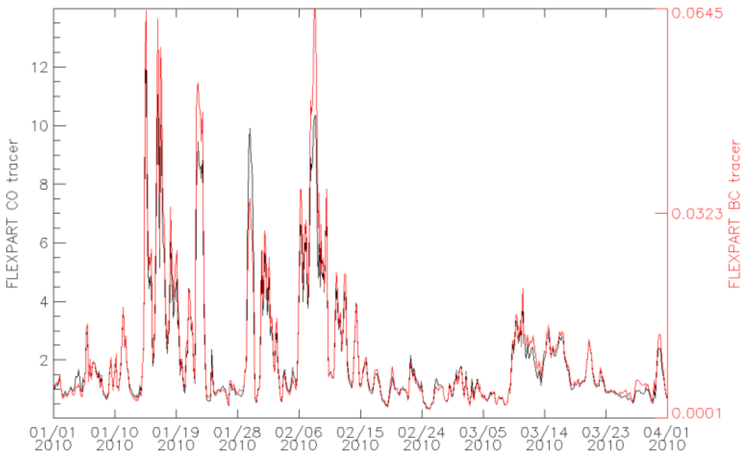

Fig R-3. Time series of the FLEXPART CO tracer (black) and the FLEXPART BC tracer (red) at Summit for Jan 01 – Mar 01 2010.

11. The retroplume altitudes as a function of plume age should be included in Figures 9, 10, and 11. Can the authors comment on the amount of NOx observed during spring and summer for observations that were likely influenced by interactions with snow (i.e. retroplumes that stay in the boundary layer for some time)?

The suggestion by the reviewer is an excellent one. The retroplumes used in this study provide an indication of the altitude where the majority of the released particles reside in 24 hour time steps (as indicated by the circles on the plots). The revised manuscript will include clearer plots and descriptions of retroplume altitudes. Retroplumes that spend a greater time at low altitudes over the Greenland ice sheet may be influenced by interactions with snowpack. These events will be identified and discussed in the revised manuscript.

12. The text describes some information, which is not adequately presented in the figures. For example, P13835 states that 42 events were identified as influenced by anthropogenic pollution using the FLEXPART BC_{anthro}. These events should be indicated on the times series of measurements (NOx, PAN, NOy, ozone) and also in Figures 5 and 6 or the time periods should be listed in the electronic supplement. Similarly, the source contribution/sensitivity to fire emissions should be shown somewhere in a figure or in the electronic supplement (similar to Figures 5 and 6, but for fires).

As suggested by the reviewer, information on the events observed, including, start time, event length and peak BC_{anthro} tracer, will be included in a table in the electronic supplement. Additionally, plots showing FLEXPART tracer contribution during fire events will be identified in a time series plot and included in the electronic supplement.

13. For the cases studied in spring and summer – how does the lifetime of PAN compare for thermal decomposition vs. photolysis (at the relevant temperatures/SZA along the trajectories)?

The primary loss process for PAN is the result of thermal decomposition. At Summit, thermal decomposition is expected to be very low as a result of the low temperatures (<0°C) throughout the year. PAN decomposition is possible during transport from lower latitudes and is expected to be the primary loss mechanism for PAN during transport to Summit. Talukdar *et al.,* 1995 showed that thermal decomposition dominates over photolysis in the low- to mid-troposphere. The retroplumes analyzed in this study indicate that pollution plumes arriving at Summit are typically transported below 6-7km and loss from photolysis is unlikely, however, it is possible that some plumes may be transported at higher altitudes before descending to Summit. The chemical transformation of the measured species during transport to and from Summit is the focus of a future study; therefore we do not feel it would be necessary to include the analyses here.

Talukdar, R. K., Burkholder, J. B., Schmoltner, A.-M., Roberts, J.M., Wilson, R. R., and Ravishankara, A. R.: Investigation of the loss processes for peroxyacetyl nitrate in the atmosphere: UV photolysis and reaction with OH, J. Geophys. Res., 100, 14163– 14173, doi:10.1029/95jd00545, 1995.

14. Is there any indication how much particulate nitrate may be contributing to NOy?

The NO_y system used at Summit is based on the same instrument design deployed at Pico from 2004-2010. Val Martin *et al.,* 2008 discussed the contribution of particulate matter to the total NO_y measured by the instrument at Pico. The authors stated that the instrument does not allow sampling of aerosols > 10 μm and although smaller nitrate aerosols can be sampled, the system used has a low conversion efficiency of these compounds. These details will be clarified in the revised manuscript.

Val Martin, M., R. E. Honrath, R. C. Owen, and Q. B. Li. "Seasonal variation of nitrogen oxides in the central North Atlantic lower free troposphere." *Journal of Geophysical Research: Atmospheres (1984–2012)* 113, no. D17 (2008).

15. The increase in the uncertainty of the PAN measurements in spring 2009 provides even further

motivation to look at the years separately. Do the increased uncertainties in 2009 impact what we can learn from the seasonal data before/after this date?

Figure R-4 shows the monthly mean PAN with the shaded region representing the uncertainty as a result of measurement and calibration uncertainty. Despite the increase in the uncertainty in spring 2009, PAN exhibits a seasonal cycle with high PAN levels in April and low levels during the summer as discussed in the manuscript. PAN measurement errors will be clarified in the revised manuscript.

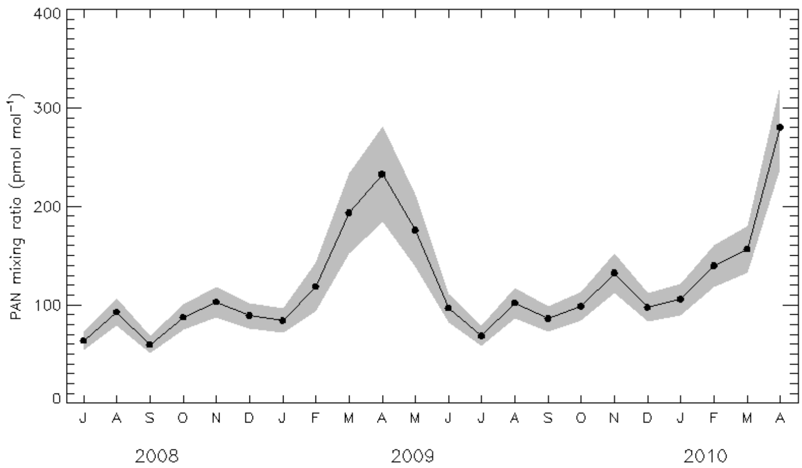

Fig R-4. Monthly mean PAN measured at Summit from July 2008 to April 2010. The shaded region represents the estimated uncertainty in the measurements.

16. A more detailed discussion of relevant POLARCAT papers is needed. Key examples include: Roiger et al., 2011 and Alvarado et al., 2010.

As suggested by the reviewer a more detailed discussion of relevant POLARCAT papers will be included in the revised manuscript.

17. Section 3.2 should be significantly rewritten/reworked. Motivation for why the seassons and events were chosen should be clearly presented at the beginning of Section 3.2. For example, the Paragraph started on line 24 Page 13834 should be presented earlier, so it's clear why the anthropogenic emissions are the focus in winter. The specific cases that are the focus of this section should be explained more clearly and the authors should endeavor to answer the question: What did we learn about anthro and BB events and their influence on NOx, PAN, and NOy at Summit by doing these measurements?

The authors agree with reviewers 1 and 2 regarding section 3.2. As discussed in the document responding to reviewer 1, a more detailed discussion on the FLEXPART source contributions and variability between the two winters will be provided at the start of section 3.2. Sub-sections 3.2.1 and 3.2.2 will be substantially re-written with additional analyses using FLEXPART retroplumes to determine the impacts of fire and anthropogenic pollution on measured species at Summit.

Technical corrections:

- Arctic should be capitalized

Corrected

- Space missing between the delta NOx and Delta ozone on page 13835 (line 24).

Corrected

- The paper should be reviewed for other typos and readability.

The revised paper will be carefully reviewed prior to submission.

References:

Roiger, A., Schlager, H., Schäfler, A., Huntrieser, H., Scheibe, M., Aufmhoff, H., Cooper, O. R., Sodemann, H., Stohl, A., Burkhart, J., Lazzara, M., Schiller, C., Law, K. S., and Arnold, F.: In-situ observation of Asian pollution transported into the Arctic lowermost stratosphere, Atmos. Chem. Phys., 11, 10975-10994, doi:10.5194/acp-11-10975-2011, 2011.

Alvarado, M. J., Logan, J. A., Mao, J., Apel, E., Riemer, D., Blake, D., Cohen, R. C., Min, K.-E., Perring, A. E., Browne, E. C., Wooldridge, P. J., Diskin, G. S., Sachse, G. W., Fuelberg, H., Sessions, W. R., Harrigan, D. L., Huey, G., Liao, J., Case-Hanks, A., Jimenez, J. L., Cubison, M. J., Vay, S. A., Weinheimer, A. J., Knapp, D. J., Montzka, D. D., Flocke, F. M., Pollack, I. B., Wennberg, P. O., Kurten, A., Crounse, J., Clair, J. M. St., Wisthaler, A., Mikoviny, T., Yantosca, R. M., Carouge, C. C., and Le Sager, P.: Nitrogen oxides and PAN in plumes from boreal fires during ARCTAS-B and their impact on ozone: an integrated analysis of aircraft and satellite observations, Atmos. Chem. Phys., 10, 9739-9760, doi:10.5194/acp-10-9739-2010, 2010.