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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal variability of atmospheric nitrogen oxides and non-methane hydrocarbons at the GEOSummit station, Greenland" *by* L. J. Kramer

Anonymous Referee #2

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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).

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?

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.

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.

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.

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.

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

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.

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.

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.

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?

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)?

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 BCanthro. 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).

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)?

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

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?

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

17. Section 3.2 should be significantly rewritten/reworked. Motivation for why the seasons 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?

Technical corrections:

- Arctic should be capitalized

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

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

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 13817, 2014.