Response to Anonymous reviewer #1

COMMENT:
This manuscript uses aircraft data and a regional chemical transport model to investigate the summertime transport of pollution from anthropogenic sources and boreal fires in North America to Greenland. The authors link aircraft observations from source regions with observations taken over Greenland to provide analysis of plume transport and plume aging. Model output is evaluated using the aircraft data then used to more broadly assess plume aging and ozone production in biomass burning versus fossil fuel plumes.

The manuscript is well written, and the material is of interest to the atmospheric composition and biomass burning communities. The central question of ozone production in boreal fire plumes is one that has been partially addressed in a number of recent studies based on the ARCTAS data. This manuscript provides clear added value in linking the near-source (ARCTAS-B) data with the downwind POLARCAT data, which have received less attention thus far. However, I have a few concerns about the statistical treatment used for assessing plume aging, the central component of the manuscript, detailed below. Once this and a few other issues are addressed, the paper is in principle suitable for publication in ACP.

RESPONSE:
The authors thank the anonymous reviewer for the careful reading of the manuscript and helpful comments. We address the specific comments in the sections that follow.

COMMENT:
1. A major focus of the manuscript is the evolution of dO3/dCO ratios from fresh to aged plumes, described in Section 5.1 and Fig. 12. The authors separate these plume types using latitude/longitude regions, then calculate the ratios for every relevant model grid cell. The results show a large degree of scatter, especially for the fresh anthropogenic plumes. Most cases shown in Fig. 12 show two distinct regimes for dO3 vs dCO. In this case, a simple linear least squares regression does not seem the appropriate tool to calculate the dO3/dCO ratio. For example, for fresh anthropogenic pollution on 1 July, this results in a ratio that isn't representative of either population of points but instead splits the difference between the two main populations. The authors follow by concluding that O3 production occurred in the plume because the slope of the best fit line changed. Looking at Fig. 12, it seems equally plausible that no O3 production occurred, but that the "aged" domain only saw one of the two initial populations on the given day. Alternatively, the "fresh" domain could include a large amount of mixed "aged" air, and the only true "fresh" plumes are those with the high dCO (relative to dO3) values shown in Fig. 12A, signifying even more substantial aging that calculated by this method.

I can think of one obvious way to improve this characterization, although there may be others. These results are based on a model run, so the exact locations of the emissions are known. Rather than using a simple lat/lon definition of fresh plumes, the calculation could be done using, e.g., only values from grid cells

containing fresh emissions, or within 2 cells of a cell with fresh emissions, or something similar. Aged plumes could still be computed based on lat/lon regions. Theoretically, this should result in less scatter in the fresh plume plots.

RESPONSE:

It is important to note that by taking differences between model runs (with-without emissions) we have already only used cells that include emissions. The question is how to separate "fresh" from "aged" plumes, which is not straightforward. For example, if we used grid cells where emissions were injected, we would limit the analysis to plumes that were only a few hours old. The focus of this study is to try and separate plumes that have aged less than a few days, from plumes that have been transported and undergone chemistry/mixing for more than 4 days (that are also transported in the direction of the Arctic). So, in order to address the reviewers comment, we have narrowed the region for what we consider "fresh" emissions. This allows for some plume processing, while significantly limiting the aging time compared to the original histograms presented in Fig. 12 of the paper.

For fires, we have taken the hourly emissions for each day and constructed a more limited lat/lon region that contains the Canadian fires. The region (which varies each day) includes only fires north of 50 °N and is limited using grid cells with emissions at least 5% of the peak emissions (see Fig. 1 below). Then, we have used this smaller region to re-calculate the distribution functions. The updated distribution functions and slopes are presented in Table 2 and Figure 12 of the paper. Figure 12 now includes both the base and FireCOSens runs, and the slopes for all best fit lines.

The changes for anthropogenic plumes are discussed in the following comment.

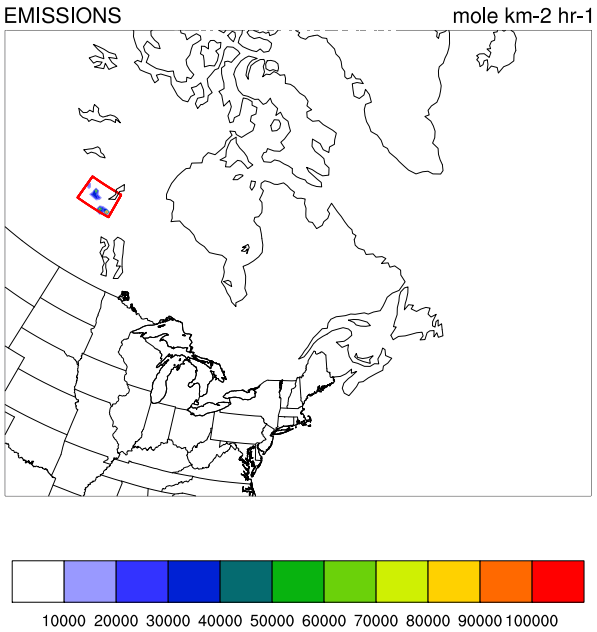

Figure 1. Example of lat/lon region (in red box) used for analysis of "fresh" plumes 1 July 2008 overlaid on fire emissions CO at 22:00 UTC on the same day.

COMMENT:
For anthropogenic plumes, an altitude cut-off may also be necessary. It wasn't entirely clear from the text, but I assume anthropogenic emissions are only emitted into the boundary layer.
RESPONSE:
Yes, that is correct – anthropogenic emissions are emitted at the surface into the boundary layer. We have limited the "fresh" emissions using lat<55 °N and added an altitude limit to include plumes <2 km for "fresh" anthropogenic plumes. The updated results are shown in Table 2 and Fig. 12 of the paper.

COMMENT:
This may mean that higher-altitude anthropogenic enrichments in source regions represent aged emissions from upwind rather than fresh local emissions, and this too could be contributing to the scatter in Fig. 12. Finally, if there is still significant scatter in these plots, it would be worth using a weighted least squares regression (for example, weighted by the probability values plotted in Fig. 12) to try to avoid the "splitting the difference" effect seen currently.
RESPONSE:
The least squared linear regression originally presented in the paper was calculated using the output of the model runs directly before calculating the distribution functions and is equivalent to a weighted least squares regression. However, due to the nature of the distribution functions, we have changed the method for calculating the slopes of the best-fit lines, as suggested by the reviewer. The method used now searches for the maximum of the distribution function for each ΔCO bin. Then, this function that represents the maximum probability for finding a specific ΔO3 value for a given ΔCO value is fit to a line. The fits are now more representative of the ΔO3 vs. ΔCO relationship. To show this we have also included additional examples in Fig. 12 of the paper to demonstrate the quality of the fits.

Given this, the conclusions based on the fits are more robust. The main change is that "fresh" plumes have more enhanced ΔO3/ΔCO fits, due to the fact that we limit the analysis to fresh emissions in the boundary layer. However, there are still moderate increases in the slope of ΔO3/ΔCO after transport. The conclusions for fires remain very similar using the new analysis method. The abstract, discussion, and conclusions have been updated to reflect the new results.

COMMENT:
2. I find the title "towards the Arctic" misleading. The majority of the model domain is sub-Arctic, the focus of the paper is on Greenland (which in summer is largely south of the polar front), and there is no discussion as to whether the observed pollution plumes are subsequently transported poleward into the Arctic or back to sub-Arctic / mid-latitude regions. This doesn't change the relevance or interest from an Arctic perspective, but rather the expectations of what the paper will contain. I would find it more accurate if "the Arctic" were replaced with "Greenland".
RESPONSE:

This has been updated and the new title of the paper is “Pollution transport from North America to Greenland during summer 2008”

Minor Comments:

COMMENT:
Page 29708, line 1: Biomass burning is not always a natural source (see e.g. spring ARCTAS papers and influence of Eurasian agricultural burning).
RESPONSE:
This is a very good point. We changed this from "natural sources such as biomass burning (BB)" to "and biomass burning (BB)" in order to not discriminate between natural and anthropogenic biomass burning.

COMMENT:
Page 29710, lines 6-10: It would be good to specify that this refers to summer POLARCAT, as a number of other CTMS have modeled ARCTAS-A data.
RESPONSE:
This has been changed to specify this refers to summer POLARCAT including ARCTAS-B.

COMMENT:
Page 29715, paragraph 2: I don’t see the value of Fig. 2. The plume transport is more clearly and relevantly demonstrated later in the paper in Figs. 9 and 11.
RESPONSE:
While the reviewer does have a point that this does not add quantitative information about the plumes to the paper, this figure does highlight the location of plumes in the model. In addition, this highlights that the plumes are at a scale that cannot be typically resolved by global models and is useful for comparing with Fig. 11. Therefore, we have chosen to leave this figure in the paper.

COMMENT:
Page 29716, lines 5-11: This doesn’t seem relevant here, unless there is a specific mismatch to be explained.
RESPONSE:
This point is to explain why this type of analysis is difficult, due to imperfect representation of plume transport in models. These sentences have been removed and the following sentence has been changed to read:
"A number of issues (resolution of plumes, imperfect representation of long-range transport, non-linear chemistry, temporal and spatial resolution of emissions) make modeling long-range transport of pollution plumes particularly challenging."

COMMENT:
Page 29716, lines 19-20: This specifies that the Figs in the SI show model results sampled along the flight tracks – are the values in Fig. 3 also sampled along the flight tracks?
RESPONSE:

Yes, the model results are extracted along flight tracks (using the flights in Table 1), then the results are used to construct the profiles. This has been clarified in the paper by adding:
"We compare vertical profiles from model results extracted along flight tracks" and later adding "(both observations and model results were binned by altitude to construct profiles)"

COMMENT:
Page 29718, lines 6-8: But decreasing NOx in this sensitivity run also decreases PAN. . . Doesn't this point to photochemistry as the more likely culprit?
RESPONSE:
By decreasing NO_x by a factor of 2, we may expect PAN concentrations that are not in agreement with the DC8 measurements in fire plumes below 2 km. However, PAN concentrations are at most decreased by 10% (Figure 3c). Therefore, there is only a small impact of decreasing the NO_x concentrations on PAN formation in the model. To clarify this, we have added:
"but has a less significant impact on ozone and PAN formation in fresh fire plumes."

COMMENT:
Page 29718, lines 17: "good" here is misleading – it would be better to just say that it's within the standard deviation (ethane looks pretty low in the boundary layer, but it's not a focus of the paper so it's not that important)
RESPONSE:
This has been changed to:
"Ethane and ethene are within the standard deviation of the measurements below 3 km, suggesting that FINNv1 captures emissions of these species reasonably well. "

COMMENT:
Page 29718, lines 19-24: Given that the aromatics can't be directly compared for the reasons stated and that they are never revisited in the paper, I don't see the value in showing and discussing them here.
RESPONSE:
The aromatics have been removed from the comparison in Figure 3, and the associated text has been removed from the paper.

COMMENT:
Page 29718, lines 25-27: This seems counterintuitive to me and requires clarification. Wouldn't a problem with fire emissions lead to worse agreement below 3km, rather than better? Unless the suggestion is that the problem is with the injection heights?
RESPONSE:
This was worded in a very confusing way and we thank the reviewer for pointing this out. The idea was to state that the agreement is much better below 3 km, where WRF-Chem injects the majority of the fire emissions. Therefore, in this region the profiles represent WRF-Chem results and not the MOZART boundary/initial conditions. Above this, where the model does not agree, the profiles are dominated by the initial conditions. The second sentence here was

intended to say that all of the issues described above, including low concentrations of VOCs are a general problem for models, and that the WRF-Chem results are not unique in the respect. These sentences have been changed to:
"We note that the agreement between the model and measurements is better below 3 km where the majority of fresh fire emissions are injected. Therefore, comparison of the profiles from 0-3 km provides the best evaluation WRF-Chem performance for fresh fire plumes. It is also important to note that the deficiencies in the representation of NMHCs and other species in models is a general problem and can be attributed to uncertainties in fire emissions inventories (e.g. Wiedinmyer et al., 2011)."

COMMENT:
Section 4 onwards: The authors make a compelling case throughout Section 3 for the FireCOSens run (2x BB CO) being a better representation of observed CO. Can that run be used as the default (improved) run in the subsequent sections and figures, rather than continually referred to as a sensitivity simulation? I think the base run could be removed from Figs. 4, 7, and 8, and the current Fig. 12 fire plots could be replaced with Fig. S10.
RESPONSE:
The strong case for the FireCOSens run being more representative of fire plumes is made in Figs. 3, 4, 7, and 8. Without the additional comparisons, especially in the ozone-co correlation plots, we don't believe it is possible to definitively conclude that this run is more representative CO values measured during POLARCAT. As suggested, we have added the distribution functions for the FireCOSens run in Figure 12.

COMMENT:
Page 29720, line 24: Why Asian emissions? These haven't been referenced elsewhere in the manuscript. Perhaps a reference to the relevance of these in the Arctic would help (e.g. Fisher et al. (2010), Shindell et al. (2008), . . .).
RESPONSE:
This is a good point. Therefore, we have added both of these references and rephrased the sentence to read:
"While the same emissions are used for both WRF-Chem and MOZART-4, the negative bias in CO may originate from differences in the model resolution and the corresponding ability to resolve plumes spatially or from Asian emissions, which may be too low in emissions inventories (e.g. Shindell et al., 2008, Fisher et al., 2010). "

Reference:
Shindell, D. T., Chin, M., Dentener, F., Doherty, R. M., Faluvegi, G., Fiore, A. M., Hess, P., Koch, D. M., MacKenzie, I. A., Sanderson, M. G., Schultz, M. G., Schulz, M., Stevenson, D. S., Teich, H., Textor, C., Wild, O., Bergmann, D. J., Bey, I., Bian, H., Cuvelier, C., Duncan, B. N., Folberth, G., Horowitz, L. W., Jonson, J., Kaminski, J. W., Marmer, E., Park, R., Pringle, K. J., Schroeder, S., Szopa, S., Takemura, T., Zeng, G., Keating, T. J., and Zuber, A.: A multi-model assessment of pollution transport to the Arctic, Atmos. Chem. Phys., 8, 5353-5372, doi:10.5194/acp-8-5353-2008, 2008.

Fisher, J. A., Jacob, D. J., Purdy, M. T., Kopacz, M., Le Sager, P., Carouge, C., Holmes, C. D., Yantosca, R. M., Batchelor, R. L., Strong, K., Diskin, G. S., Fuelberg, H. E., Holloway, J. S., Hyer, E. J., McMillan, W. W., Warner, J., Streets, D. G., Zhang, Q., Wang, Y., and Wu, S.: Source attribution and interannual variability of Arctic pollution in spring constrained by aircraft (ARCTAS, ARCPAC) and satellite (AIRS) observations of carbon monoxide, Atmos. Chem. Phys., 10, 977-996, doi:10.5194/acp-10-977-2010, 2010.

COMMENT:
Page 29721, lines 6-14: What would the equivalent of the purple trace in Fig. 6C look like for NO, O3, and PAN? In other words, does the plume offset seen from the CO affect the profiles of all species, or just CO?
RESPONSE:
The profiles for the other species at the same time/location are shown below in Fig. 2. These results highlight that, in this particular case, the troposphere extends to a higher altitude than the campaign average resulting in lower ozone mixing ratios in the region above 8 km. A PAN enhancement is co-located with the CO plume, but this plume has lower NO mixing ratios than the campaign average. We have highlighted in the paper that for this plume, the model enhancement is not co-located with the aircraft in the model, which is one of the challenges of evaluating higher resolution chemical transport models.

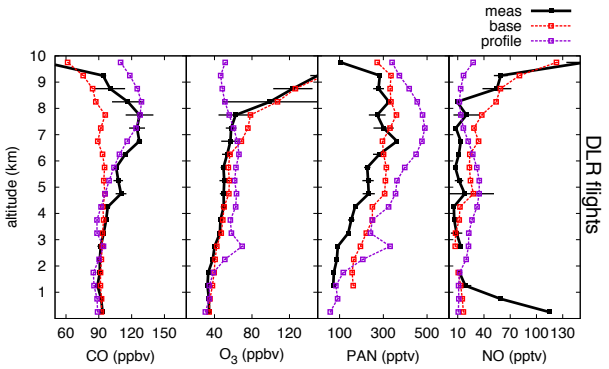

Figure 2. Vertical profiles for CO, ozone, PAN, and NO extracted from the model run (grid cell center = 61.1˚N, 41.5 ˚W) on 8 July at 12:00 UTC compared to profiles constructed from the Falcon-20 flight tracks using measurements (black) and the base case model run (red).

COMMENT:
Page 29721, lines 11-12: Some comment is needed on the large model-observation discrepancy in NO below 1.5 km shown in Fig. 6F.
RESPONSE:
The very high NO values below 1 km are local airport pollution, which is not included in the emissions inventory. A statement noting this has been added to the paper.

COMMENT:
Page 29722, line 24: Is the offset of 15 min in the model significant? What is the model time step? If this is only an offset of 1 time step it may be worth mentioning (since that is still good agreement).
RESPONSE:
The model time step used is 3 minutes for dynamics, therefore this represents several model time steps. This represents a spatial displacement of the plume in the model, which occurs two model grid cells away from where it was seen by the aircraft. The spatial displacement of the plume is approximately 70 km, which is not surprising considering the duration of the WRF run. We have added the text:
"representing a spatial displacement of the plume of 70 km in the model from where it was seen by the aircraft. This spatial displacement of the plume represents very good agreement for an aged plume that has undergone long range transport in a high resolution model run."

COMMENT:
Page 29733, lines 4-6: The finding that boreal fire CO is too low in FINNv1 shouldn't be dismissed a caveat – this is an important result for the community and should be highlighted as such!
RESPONSE:
We have added "an important conclusion regarding the CO emissions in the FINNv1 emissions inventory".

COMMENT:
Page 29733, lines 7-8: In Section 3.1 it was argued that the high NOx may reflect problems with the photochemistry (conversion from PAN), not necessarily the fire emissions as blamed here. . .
RESPONSE:
This can be due to emissions, photochemistry, or a combination of the two. Therefore, we have removed the phrase attributing this to only the emissions inventory.

COMMENT:
Page 29733, lines 24-26: The authors also found that these two contributions were not mixed – this is an important result and is worth highlighting here.
RESPONSE:
The reviewer is right that most of the examples highlighted in the text are not mixed plumes. However, there are cases shown where these contributions are mixed, for example the contribution to ozone mixing ratios in Figure 8D (flight leg at 7 km). To address this we have added:
"with the majority of plumes analyzed consisting of either anthropogenic or BB pollution, rather than mixtures."

Technical Comments:

COMMENT:
Page 29715, lines 13-14: As written, this suggests that fires "sampled by the DC-8" were only part of the "model run" but they were also clearly observed. . . Consider replacing "model run" with "observation period" or something of that nature.
RESPONSE:
This was changed to:
"The observation period and model run include intense boreal forest fires over Canada that were sampled by the DC8"

COMMENT:
Page 29716, lines 20: I suggest changing "all" to "individual" – as is, it sounds like Fig. 3 only contains some of the flights.
RESPONSE:
This has been updated as suggested.

COMMENT:
Page 29723, lines 6-10 and Page 29725, lines 8-14: The discussions of high O3 / low CO are confusing where they are and would be better left to the later discussions of the lidar measurements and stratospheric folds.
RESPONSE:
We have moved the majority of this discussion to the section on the lidar measurements.

COMMENT:
Page 29727, line 3: Please add a reference to the more recent Akagi et al. (2011).
RESPONSE:
This reference has been added, we thank the reviewer for catching this oversight.

Reference:
Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D., and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmos. Chem. Phys., 11, 4039-4072, doi:10.5194/acp-11-4039-2011, 2011.

COMMENT:
Page 29732, lines 11-16: This is very repetitive of the previous sentences and can probably be cut.
RESPONSE:
These sentences have been removed.

COMMENT:
Page 29733, line 8: I would replace "French and German" with "POLARCAT" (or remove altogether) – as is this will be confusing for a reader who doesn't read the details of the campaigns and who organized them.
RESPONSE:
This has been replaced as suggested.

COMMENT:
Page 29733, lines 11-14: I suggest removing this sentence, which isn't really a finding of the paper and doesn't add anything to the other substantial conclusions presented here.
RESPONSE:
These sentences have been removed as suggested.

COMMENT:
Page 29734, line 5: Reference(s) needed here
RESPONSE:
We have added a reference to Alvarado et al., 2010.

Reference:
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.

COMMENT:
Figs. 3, 5, 6, and 8 would each benefit from having a legend on the figure itself (not just in the caption).
RESPONSE:
Legends have been added to these figures.

COMMENT:
Fig. 7: It is hard to qualitatively compare these plots. As commented previously, I think you could remove the "Base" plot and just show "FireCOSens". It also might be nice to show some sort of statistical measure of agreement here (e.g. the model-observation correlation coefficient for the O3/CO ratios shown in this plot), especially if you decide to keep both sets of model plots, to show which is "better."
RESPONSE:
Box and whisker plots for data along flight tracks for the measurements, base, and FireCOSens runs have been added to Figs. 4 and 7.

COMMENT:
Fig. 8: The gray is hard to see (not visible at all on my printer) – can it be darkened somewhat?
RESPONSE:
Both the gray and yellow color have been make darker, thanks for pointing this out.

COMMENT:
Fig. 10: It would be better if the format of the time (x) axis matched the format used in the text (e.g. 13:30 instead of 13.5, etc.).
RESPONSE:
This has been updated in the new figure.

COMMENT:
Fig. 11: It would be valuable to also show maps of dCO here, to support the discussion on the relative dO3 vs dCO in plumes (visual example of O3 increase during transport relative to CO).
RESPONSE:
The ΔCO values are very similar to the plots already shown in Figure 2 for the base run (Figure 2d and 2e), therefore we have left Figure 2 in the paper and added a reference to these plumes in the Figure 11 caption.

COMMENT:
Fig. 12: The colorbar is missing an axis label.
RESPONSE:
This value is unitless, because the distributions are normalized to the number of grid cells. Therefore we have not added a legend to the label bar.