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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Pollution transport towards the Arctic during summer 2008" *by* J. L. Thomas et al.

Anonymous Referee #1

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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.

Major Comments

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. 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. 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.

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".

Minor Comments

Page 29708, line 1: Biomass burning is not always a natural source (see e.g. spring ARCTAS papers and influence of Eurasian agricultural burning).

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.

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.

Page 29716, lines 5-11: This doesn't seem relevant here, unless there is a specific mismatch to be explained.

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?

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?

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)

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.

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?

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.

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), ...).

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?

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.

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).

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!

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. . .

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.

Technical Comments

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.

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

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.

Page 29727, line 3: Please add a reference to the more recent Akagi et al. (2011).

Page 29732, lines 11-16: This is very repetitive of the previous sentences and can probably be cut.

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.

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.

Page 29734, line 5: Reference(s) needed here

Figs. 3, 5, 6, and 8 would each benefit from having a legend on the figure itself (not just in the caption).

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."

Fig. 8: The gray is hard to see (not visible at all on my printer) – can it be darkened somewhat?

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.).

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).

Fig. 12: The colorbar is missing an axis label.

References

1. Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D., and Wennberg, P. O.: Emis- sion 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.

2. 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 air craft (ARCTAS, ARCPAC) and satellite (AIRS) observations of carbon monoxide, Atmos. Chem. Phys., 10, 977–996, doi:10.5194/acp-10-977-2010, 2010.

3. 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.

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