Arnold et al. – "Biomass burning influence on high latitude tropospheric ozone and reactive nitrogen in summer 2008: a multi-model analysis based on POLMIP simulations".

Response to Reviewer comments

We thank the two anonymous referees for their careful review of our manuscript. Here we respond to comments of Reviewer 2. Reviewer comments are given in italic text, and our response in normal text. We have shown in bold new text that we have added to the manuscript in response to the comments.

Anonymous Referee #2

- There is no mention of how photolysis is treated in the models. Certainly radiation-driven processes are important in the summer Arctic troposphere, and it would be good to describe how this is simulated in the models. Are "real-time" clouds, aerosols, surface albedos affecting the simulation? And how do factors such as the above behave during the period of study in that location (Mostly cloudy conditions or not? Air masses passing over ice? etc). Ideally there should be some analysis of the photolysis rates themselves - in addition to the concentrations and chemical fluxes - but if that is not possible, at least some discussion would be useful to the reader.

We did not present a discussion and evaluation of photolysis in the models, since this is presented in the POLMIP overview paper (Emmons et al., 2014). This paper was submitted and appeared in ACPD some time after submission of our manuscript, so its content may have been unavailable to the reviewer. Briefly, Emmons et al., (2014) describe the treatment of photolysis in the models, show differences between model cloud fractions and $J(O^1D)$ and $J(NO_2)$ photolysis rates, and compare these with ARCTAS data from the NASA DC8 aircraft. We do not feel that it is worthwhile repeating this analysis here. However, we have added text in the penultimate paragraph of Section 3.2 to explicitly highlight that differences in simulated photolysis rates likely contribute to differences in ozone production efficiency relative to high latitude CO enhancement:

"**Differences in simulated photolysis between the models are likely contributors to model spread in photochemical ozone enhancement relative to CO. Such differences are presented and explored for the POLMIP models by Emmons et al., (2014).**"

- Ozone-CO correlations/slopes can be very useful, but being positive is not always indicative of net ozone producing regions, especially over remote areas (as suggested by Voulgarakis et al. (2011) and later also discussed by Kim et al. (2013) and Zhang et al. (2014)). Examining the OH levels in comparison to the background and subsequent CO destruction fluxes would give an indication on the validity of this approach.

This same point was also raised Reviewer 1. We have addressed the point by evaluating the simulated photochemical loss rate of CO in the air masses and compared it with the photochemical ozone production tendency. See our response to Reviewer 1 above.

MINOR COMMENTS:
Page 24580, Lines 6-7: A little more justification of the choice of a 25-day lifetime is needed, for the more general readership.

The 25-day lifetime is somewhat arbitrary, but has been chosen in order to produce tracers with an atmospheric lifetime that is sufficiently long compared with characteristic transport timescales for long-range transport from source regions to the Arctic (a few

days to a week), but short enough to avoid a homogeneous well-mixed abundance accumulating during the simulation. A lifetime of 25 days is shorter than the expected range in lifetime of CO corresponding to a range in tropospheric [OH] of between 1.0 $x10^6$ and $2.0x10^6$ molec cm^{-3} (roughly 35-80 days). We have added the following sentence to the manuscript: **"A 25-day tracer lifetime is sufficiently long relative to the transport timescale for long-range transport from mid-latitudes to the Arctic (days to a week), while being short enough to avoid the formation of a homogeneous well-mixed tracer distribution."**

Figures 1-4: It would be good to clearly label the axes, i.e. which one is the observations and which one is the model?

Extra labels have been added to more clearly designate model and observations in these figures.

Page 24582, Lines 4-6: Worth mentioning that models typically underestimate CO in the northern extratropics, e.g. see Naik et al. (2013), Fig. 2 for a recent multi-model comparison.

We have added a short section to the text: "**Global models typically underestimate CO in the northern extratropics. A recent multi-model study showed negative annual mean model biases exceeding -45 ppbv compared with surface CO observations at high latitudes, and as large as -30 ppbv compared with satellite-retrieved CO concentrations at 500 hPa over the extra-tropical oceans (Naik et al, 2013)."**

Page 24583, Line 4: Please spell out "oVOC" as it is the first time it is encountered in the text.

Done. Also removed redundant spelling out of oVOC later in manuscript (page 24594, line 12).

Page 24583, Lines 4-7: Any ideas on why oVOCs show such a large variability? Is it a result of different emissions, or of atmospheric processing?

Emissions of these species (acetaldehyde, acetone) are the same for all models – with the exception of some differences for GEOS-Chem and TM5 (see Table 2 in Emmons et al., 2014). The large diversity between the majority of models is therefore a result of differences in photochemical production of these species, due to both differences in treatments of organic chemistry and differences in rates of photochemical processing of parent VOCs that go on to produce these species, as well as differences in their photolysis and OH loss. We have added the following text to the paper to point this out explicitly: **"With the exception of the GEOS-Chem and TM5 models, emissions of acetone and acetaldehyde are the same for all models. The large diversity in model concentrations of these species therefore mainly results from different treatments of organic chemistry, differences in rates of photochemical processing of their parent VOCs and differences in their photolysis and OH loss."**

Figure 4: For SMHI-MATCH the bias appears positive (95%), but visually the figure suggests a negative bias.

The percentage figures quoted in the panels of Figures 1-4 were mean bias. These values were therefore heavily weighted by any points with particularly large bias, even if there are few of them. This was the case for the SMHI-MATCH model in this figure

where the maximum bias is 55,000%, leading to a positive mean bias despite most points having a negative bias. We have modified the plots to show median bias instead, to remove this effect.

Figure 5: "(k)" should not be bold.

Corrected.

Figure 6: I would suggest using consistent colouring for young/aged in Figures 5 & 6.

Agree with this suggestion. We have adjusted the colours used in Figure 6 to be consistent with Figure 5.

Figures 5 & 7: In those figures, some model names are different to previous figures.

We have amended model names used, and checked that these are consistent throughout the manuscript.

Page 24587, Lines 22-24: I presume the authors here imply how future model developments regarding convection could affect the results. This should be more clearly stated.

No – this statement refers to the implementation of convective vertical transport in the models as used in the study. The point being made is that during a period of increased convective activity, vertical redistribution of tracer due to convection may partially mask any differences in tracer vertical profile produced by differences in large-scale resolved vertical motion in the driving meteorological data. In many global models, such convective transport is parameterised. We have modified the statement to make its meaning clearer: **"Increased convective vertical mixing in the models may therefore mask some of the differences in vertical tracer structure produced by differences in large-scale vertical transport."**

Page 24589, Line 13: Please change "represented" to "be represented".
Done.

Page 24590, Lines 10-13: It is not clear to me that this is the case. E.g. the CIFS model looks much more similar to TM5 next to it or CAM5 above it rather than to SMHI-MATCH.
This statement refers specifically to the sharpness of gradients in the CO tracer structure close to source regions, and not to tracer concentration values. While the concentration values for CIFS appear more similar to TM5 and CAM5, at least visually there is evidence that there are sharper gradients in filamentary structures close to the E Asia coast (e.g. over the Sea of Okhotsk, to the north of Japan) in the CIFS model on a comparable scale with those in SMHI-MATCH. However, in our analysis we have not made a quantitative assessment of the comparability of these structures and gradients. We feel therefore that this statement was perhaps overly subjective, and so have removed the sentence in question from the manuscript. This does not at all change the key aspects of our discussion or conclusions of our results.

Table 2: It would be useful to show OH on this table too, in order to get a sense of the variability between the models.

OH has been added to the table as suggested.

Page 24594, Lines 13-18: Is the factor of 2 arbitrary or based on the typical diversity range in the POLMIP models (e.g. from Emmons et al., 2014)? Please clarify.

The choice of a factor 2 is somewhat arbitrary, but is consistent with the magnitude of inter-model diversity shown for several species (PAN, oVOCs) along the DC8 flight tracks in Emmons et al., (2014). We chose to apply the same fractional perturbation to each species to allow direct comparison of the relative sensitivity of ozone production their abundances. We have modified the text to clarify this point: **"A factor 2 perturbation is consistent with inter-model differences and biases along the ARCTAS DC8 flight tracks (Emmons et al., 2014). We apply the same fractional perturbation to each species to directly compare sensitivities of Arctic ozone photochemistry to uncertainties in their abundances."**

Page 24595, Lines 3-6: I am not sure I understand, though I may be missing something here: All the lines in Figure 14c seem to be below the zero line, so I am not sure where one can see an enhancement of ozone.

We apologise, as the intended meaning of this text is somewhat unclear and/or misleading. The point is that e.g. the negative perturbation to oVOCs produces an enhancement to ozone relative to the control case with unperturbed oVOCs, which is demonstrated by a smaller net loss in ozone over the 4-day period – this is not necessarily an enhancement in ozone relative to initial ozone concentration. We have reworded this sentence to improve clarity of intended meaning: **"Increasing and decreasing initial oVOC abundances leads to enhancement and suppression of ozone loss in the plume respectively over the following 4 days (Fig. 14c and d), due to the role of acetaldehyde and acetone as a source of the peroxyacetyl radical during their photo-oxidation."**

Page 24595, Line 16: Please change "changes the rate" to "changes of the rate".

Done.

Page 24596, Line 11: Please change "differences efficiency" to "differences in the efficiency".

Done.

References

Emmons, L. K., Arnold, S. R., Monks, S. A., Huijnen, V., Tilmes, S., Law, K. S., Thomas, J. L., Raut, J.-C., Bouarar, I., Turquety, S., Long, Y., Duncan, B., Steenrod, S., Strode, S., Flemming, J., Mao, J., Langner, J., Thompson, A. M., Tarasick, D., Apel, E. C., Blake, D. R., Cohen, R. C., Dibb, J., Diskin, G. S., Fried, A., Hall, S. R., Huey, L. G., Weinheimer, A. J., Wisthaler, A., Mikoviny, T., Nowak, J., Peischl, J., Roberts, J. M., Ryerson, T., Warneke, C., and Helmig, D.: The POLARCAT Model Intercomparison Project (POLMIP): overview and evaluation with observations, Atmos. Chem. Phys. Discuss., 14, 29331-29393, doi:10.5194/acpd-14-29331-2014, 2014.

Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J.-F., Lin, M., Prather, M. J., Young, P. J., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R., Eyring, V., Faluvegi, G., Folberth, G. A., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., van Noije, T. P. C., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R., Shindell, D. T., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., and Zeng, G.: Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model

Intercomparison Project (ACCMIP), Atmos. Chem. Phys., 13, 5277-5298, doi:10.5194/acp-13-5277-2013, 2013.