

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 made by Reviewer 1. 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 #1

My main concern is the use of O₃/CO as a diagnostic for ozone production. Recent work (e.g. see Zhang et al. [2014]) advises against using this diagnostic without verifying the assumption of negligible CO loss. Could the authors do that prior to publication in ACP? That would provide a much stronger foundation for the remainder of the analysis.

In order to address this point, we have investigated the chemical loss rate of CO (due to CO+OH reaction) in the region of our analysis (latitude > 50N, 850-250 hPa) in the MOZART-4 model. This model has supplied output on chemical rates from the POLMIP simulation. We find that daily chemical loss rates of CO are small (average 1.9 ppbv/day), ranging between 1.5%-4.5%, and are also partly compensated by chemical production of CO from VOC oxidation (average 0.9 ppbv/day) at the same locations. We compared the chemical CO loss rate with the chemical production rate of ozone at each model grid-point in the specified region. This comparison demonstrates that the chemical loss of CO is negligible, and is substantially outweighed by the fractional photochemical production rate of ozone in these air masses. We believe that this analysis provides a sound basis for our assumption that the ozone/CO slopes are useful diagnostics of chemical ozone formation. We have included some text in the manuscript to summarise our justification for the use of ozone/CO slopes: **“Recent studies have highlighted the need for caution regarding the use of O₃/CO slopes to diagnose photochemical ozone production, particularly in remote regions, due to slopes being artificially increased by chemical loss of CO due to reaction with OH (e.g. Voulgarakis et al., 2011; Zhang et al., 2014). Chemical rate output from the MOZART-4 model shows that in the domain of our study (latitude 50N-90N, 850-250 hPa) the daily chemical loss rate of CO is small (average 1.9 ppbv/day), equivalent to 1.5%-4.5%. This loss is partly offset by chemical production of CO from VOC oxidation (average 0.9 ppbv/day), and daily fractional rates of chemical ozone production at the same locations are substantially larger (~5 - 45%). This analysis suggests that chemical CO loss is unlikely to have a significant effect on our calculated ozone/CO slopes.”**

Pg: 24575, lines 12-13: Grammar issue here. Missing “in”

Corrected.

Pg:24584, lines 25 onward: This designation would still be problematic if there was CO loss in the plume.

We have addressed the issue of CO loss above, and in light of our diagnosis of negligible CO loss we retain this designation and discussion.

Pg: 24586, lines 7-9: The authors should describe a) the implementation of and rational

for the HO₂ uptake in GEOS-Chem, and 2) how this implementation impacts the abundances of PAN and other species relevant to the paper.

Uptake of HO₂ is included in GEOS-Chem such that it is irreversibly lost through conversion to H₂O on aerosol, rather than forming H₂O₂. The motivation for this is that joint atmospheric observations of HO₂ and H₂O₂ from field studies suggest that HO₂ uptake by aerosols may not produce H₂O₂. It has been proposed that the conversion of HO₂ to H₂O can be catalyzed by transition metals in aqueous aerosols. This motivation and implementation of the process in GEOS-Chem has been described and implemented in the GEOS-Chem model (details in Mao et al., (2013)). We do not have a comparable simulation without this treatment of HO₂ uptake with which we can compare PAN and other NO_y species for POLMIP. Results from the Mao et al., (2013) study showed that in simulations with the HO₂ → H₂O aerosol conversion included, HO_x concentrations are reduced due to reduction in the re-formation of HO_x via photolysis of H₂O₂ (a 12% reduction in mass-weighted global mean OH). This was shown to increase CO concentrations due to an increased CO lifetime, particularly in the extra-tropics. In addition, surface ozone concentrations decrease in general by 3–10 ppb over North America and Eurasia relative to a simulation without the HO₂ → H₂O uptake. We have added text to Section 2 to summarise these points: **“The GEOS-Chem model includes a parameterization for transition metal-catalysed formation of H₂O from aerosol uptake of HO₂, rather than formation of H₂O₂. This process is effectively an irreversible loss for HO_x, and is motivated by the suggestion from field observations that HO₂ uptake to aerosol may not produce H₂O₂. This motivation and the implementation of this scheme are described by Mao et al., (2013). The same study showed that inclusion of this process reduces the mass-weighted global mean OH concentration by 12%, and substantially increases CO concentrations at high latitudes due to an increased CO lifetime. It was also shown to reduce surface ozone by 3-10 ppbv over North America and Eurasia.”**

Pg: 24588: lines 19-22: The standard version of GEOS-Chem does not emit NO_y with this partitioning. This indicates that the model used in the comparison should probably be better documented. Somewhere in the text should point to a reference for this version with a statement that the model is not a public release version.

We apologise, but the information provided on NO_y partitioning does not in fact apply to the POLMIP GEOS-Chem simulations. For implementation of the FINN fire emissions, the emissions were speciated as NO_x only, to be consistent with the other models. We have removed the information on modified emitted NO_y partitioning from the manuscript.

Figure 5 and Figure 7: There seem to be excessive significant figures in this set of figures.

Agreed. We have reduced these to 3 significant figures, consistent with their presentation in the main text.

References

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