

## ***Interactive comment on* “Sources contributing to background surface ozone in the US Intermountain West” by L. Zhang et al.**

**L. Zhang et al.**

linzhang.atm@gmail.com

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**Comment:** This analysis provides incremental improvements to the GEOS-Chem simulations for surface ozone in the western US for the period 2006–2008, as described previously (Zhang et al 2011). The model improvements include improved lightning emissions and a daily emission inventory for wildfires. The changes in lightning emissions appears to make a significant improvement over the earlier work, especially for the southwestern US. However overall, it is not clear if the model is improved overall, it seems like it must be. It would be helpful to state this explicitly in reference to Figure 5 (eg what is the bias and r value from the earlier work?).

As for the changes in biomass burning impacts, it is not clear whether the daily inven-

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tory results in any significant improvement. The inventory is still computed on a 1x1 degree grid, which is far too coarse to adequately represent wildfires. The emissions appear to ignore influence from flaming vs smoldering combustion, which can occur in the same region at the same time. The chemistry simulation appears to be quite incomplete and relies on unrealistic tricks, such as emitting PAN directly with other primary emissions. How can this accurately simulate the chemistry? It is not surprising, then, that the model fails to reproduce the observations in fire impacted regions. The large over-estimate in ozone production shown (Figure 8) likely reflects a combination of too much NO<sub>x</sub> and instantaneous dilution into a 1x1 degree grid cell. The Glacier NP case shown, clearly shows evidence of NO<sub>x</sub> titration that is not evident in the model. Yet, the authors use the model failure here, to argue that wildfires never produce ozone. Seems like an odd argument and it's directly counter to dozens of papers that show wildfire ozone production from measurements. I certainly agree that the ozone production from wildfires is complex and not fully understood. In that light, I think it would be more instructive to find cases where observations demonstrate ozone productions from wildfires and then examine the model behavior for those cases.

Overall, this paper is a useful, if incremental, improvement in our understanding of ozone. I believe the changes in the lightning analysis is the most robust and useful contribution. The improvements in the model calculated wildfire influence do not seem as robust. Other comments below:

**Response:** We thank the reviewer for the helpful comments. All of them have been implemented in the revised manuscript. To address the reviewer's first comment on the model improvement, we compared Figure 5 of this work to that in our earlier work (Zhang et al., 2011). We now state in the text "Previous model comparison with the same CASTNet observations for summer 2006 showed a relatively low correlation ( $r = 0.30$ ; Figure 3 of Zhang et al. (2011)) and high biases of 12% in the southwest US (Zhang et al. (2011)). The summertime comparison in this work ( $r = 0.65$ ) is significantly improved relative to Zhang et al. (2011) due to the modifications to lightning

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emissions as further discussed below.”

We have reorganized and rewritten Section 3.2 of the manuscript to discuss uncertainties in the wildfire emissions and in the model chemistry. Please see our itemized responses below. They also addressed the comments above.

**Comment:** P 25872, Line 10: The statement about CASTNET observations not showing evidence for O<sub>3</sub> production is a bit limiting. Why don't you look at one of the dozens of papers where observations show O<sub>3</sub> production. This statement is at odds with an extensive literature on O<sub>3</sub> from wildfires.

**Response:** We revised the sentence in the abstract to “Wildfire plumes generate ozone events in excess of 80 ppbv in GEOS-Chem, but CASTNet ozone observations in the Intermountain West show no apparent enhancements during these events nor do they show evidence of regional fire influence. Models may overestimate ozone production in fresh fire plumes because of inadequate chemistry and grid-scale dilution.”

**Comment:** 25876, line 26: The 1x1 resolution for the fire emissions seems to be quite a problem, since most wildfires will be much smaller. How does this compare to typical fire size?

**Response:** We now state in Section 3.2 “Model overestimate of ozone production in fresh fire plumes may result from inadequate chemistry, uncertainties in the emission factor and coarse grid resolution. Typical fire size recorded in the fire reports ranges 2-50 ha and large fires are over 400-10<sup>4</sup> ha, which are much smaller than the model 1/2° × 2/3° horizontal resolution.”

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**Comment:** 25881, line 20: Others have suggested that PAN is central to the ozone production. Can you describe how PAN is produced in GEOS-Chem and whether this version incorporates any changes to the PAN chemistry that I have seen discussed by E.Fischer?

**Response:** We have rewritten the section for discussing the role of PAN on ozone production in fire plumes. The discussion now reads:

“Ozone production in fresh plumes is limited by fast conversion of NO<sub>x</sub> to peroxyacetyl-nitrate (PAN) (Jacob et al., 1992; Alvarado et al., 2010), but subsequent decomposition of PAN in aged plumes could lead to ozone enhancements far downwind (Jaffe and Wigder, 2012). This effect could be magnified by buoyant plume lofting above the boundary layer, followed by ozone production over an aging time of a few days. There are many observations of elevated ozone in aged fire plumes sampled from aircraft and at mountain sites (Mauzerall et al., 1998; Real et al., 2007; Jaffe and Wigder, 2012). These plumes could then be fumigated to the surface by boundary layer entrainment and cause high ozone in surface air.”

“We use the standard GEOS-Chem chemical scheme as described by Horowitz et al. (1998), Bey et al. (2001) and Mao et al. (2010), and find little PAN-driven regional production of ozone from fires in GEOS-Chem, as reflected by the small contribution from wildfires to ozone over the scale of the Intermountain West (Figure 8). A recent study with an improved PAN chemistry also showed that fires in GEOS-Chem make little contribution to PAN at northern mid-latitudes in summer (Fischer et al., 2013). However, this could be because the model does not account for very short-lived VOCs emitted by fires and cannot resolve photochemistry on the scale of the fire plumes.”

Add reference: Fischer, E. V., Jacob, D. J., Yantosca, R. M., Sulprizio, M. P., Millet, D. B., Mao, J., Paulot, F., Singh, H. B., Roiger, A. E., Ries, L., Talbot, R. W., Dzepina, K., and Pandey Deolal, S.: Atmospheric peroxyacetyl nitrate (PAN): a global budget and source attribution, *Atmospheric Chemistry and Physics Discussions*, 13, 26841–26891,

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10.5194/acpd-13-26841-2013, 2013.

Real, E., Law, K. S., Weinzierl, B., Fiebig, M., Petzold, A., Wild, O., Methven, J., Arnold, S., Stohl, A., Huntrieser, H., Roiger, A., Schlager, H., Stewart, D., Avery, M., Sachse, G., Browell, E., Ferrare, R., and Blake, D.: Processes influencing ozone levels in Alaskan forest fire plumes during long-range transport over the North Atlantic, *J. Geophys. Res.*, 112, 10.1029/2006jd007576, 2007.

**Comment:** 25882, line: I agree that the value of 3 g/NO per kg fuel is too high. But note that it is also highly variable from fire to fire. You should discuss this variability. What would you expect for a RANGE of emission factors across the region? Certainly this is a significant limit to the model simulations.

**Response:** We now state in the text “Akagi et al. (2011) summarized recent emission factor measurements and recommended a mean value for extratropical fires of 1.12 g NO per kg dry mass burned. However, the NO<sub>x</sub> emission factor varies significantly spatially and temporally depending on the local combustion efficiency (smoldering vs. flaming) and biomass nitrogen load (Jaffe and Wigder, 2012).”

**Comment:** 25882, line 8: I am not convinced that a model run with such unrealistic emissions (PAN, HNO<sub>3</sub>) tells us anything about O<sub>3</sub> chemistry. O<sub>3</sub> production will depend critically on the HO<sub>x</sub>/NO<sub>x</sub>/RO<sub>x</sub> chemistry in the plume, so emitting these species directly seems to me to be too unrealistic to provide anything useful.

**Response:** We now state in this section “Direct emission of fire NO<sub>x</sub> as PAN in the model is likely inadequate as the PAN can decompose back to NO<sub>x</sub> in the absence of supporting VOCs and with rapid dilution on the grid scale. A Lagrangian plume-in-grid approach may be needed, such as has been implemented in GEOS-Chem for ozone production in ship plumes (Vinken et al., 2011)”. Although this is not what emissions

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occur in reality, we think it can provide a valuable test for Eulerian models to represent the fast conversion of NO<sub>x</sub> to PAN in fresh fire plumes.

**Comment:** 25882, line 26 and following paragraph: The hypothesis that the large scale temp-fire-O<sub>3</sub> correlations is related to BL height is interesting and plausible, but certainly not proven. It is going to be very challenging to untangle these comingled variables. While the GEOS-Chem simulations provide some helpful clues, given the weakness with the Chemistry simulations, I don't think we can discard any of these hypotheses at this time.

**Response:** We now state in the paragraph "As discussed above, the model may not capture the possible regional ozone enhancements from transport of PAN in fire plumes. Further observational evidence is needed to address the issue."

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 25871, 2013.

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