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Atmospheric Chemistry and Physics Discussions

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

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Comment: This paper uses a high-resolution version of the GEOS-Chem model and data from the EPA CASTNet ozone monitors to quantify the contribution of lightning, wildfires, stratospheric intrusions, and California anthropogenic pollution on background surface ozone in the US Intermountain West. This study uses an updated estimate of lightning NOx emissions based on the National Lightning Detection Network (NLDN) and a daily estimate of wildfire emissions built from fire reports from the natinal Fire and Aviation Management WEB (FAMWEB). The authors find that their improved lightning NOx estimates corrects previous ozone overestimates over the Southwest US in summer, and that lightning results in a mean 10 ppbv enhancement of ozone in

the intermountain west. They find that stratospheric intrusions are responsible for the highest ozone concentrations observed at CASTNet sites, and that while GEOS-Chem underestimates the impacts of stratospheric intrusions on surface ozone, this bias is predicatable. Differences in stratospheric influence between Zhang eta l. (2011) and Lin et al. (2012) are mostly due to differences in definition of "stratospheric influence." Finally, the authors find that while GEOS-Chem predicts large enhancements of ozone from wildfires, the CASTNet data does not show any corresponding increase. The authors suggest that previous correlations between CASTNet ozone and fire occurance are due to the correlati0on of both with surface temperature, rather than a causal relationship.

This is a well-written paper on an important topic, the methods used are reasonable, and the conclusions are generally well-supported. The new techniques used for lightning NOx emissions clearly improve model performance at selected sites. I have a few questions about the methods and results, as detailed below, but nothing serious enough to prevent publication. I think the paper should be accepted following minor revisions to address the issues listed below.

Response: We thank the reviewer for the helpful comments. All of them have been addressed in the revised manuscript. Please see our itemized responses below.

Comment: L19, P25876: On my first read through, it wasn't clear where the reduction in NOx yield per flash came from, but I'm now assuming that is due to the change in the location of the tropics/extratropics boundary? If so, please make that clearer in the text.

Response: Yes, we now add here "(due to moving the tropical/extratropical boundary for NOx yields as described above)".

Comment: L8-9, P25877: I'm not sure this is correct. I think GFED2 only gives the total carbon burned/emitted, and then emission factors for specific trace gases and particle types (CO, NOx, VOCs, OC, BC, etc.) are taken from literature reviews like Andreae Merlet (2001) or Akagi et al. (2011). What was the EF source for this study?

Response: We corrected in the text "Fuel consumption rates are taken from GFED2, and the emission factors of gases and aerosols are from Andreae and Merlet (2001) as in GFED2."

Comment: L12-14, P25877: Is this because GFED2 is missing small fires? Is there evidence from Yue et al. (2013) or elsewhere that this daily inventory is more accurate than GFED2 for the US from 2006-2008?

Response: We added in the text "GFED2 uses area burned products from the MODIS satellite instrument at 500m resolution (van der Werf et al., 2006), and thus generally misses small fires with area burned smaller than 25 ha, but those small fires account for only 0.6% of the total areas burned in the fire reports for summer 2006-2008. The difference between GFED2 and fire reports is thus mainly due to relatively large fires."

Comment: L21-25, P25877: Did Barrett et al. (2012) use the same meteorology and other relevant model settings as in this study?

Response: We state in the text "Barrett et al. (2012) tested vertical transport in GEOS-Chem with GEOS-5 meteorological data and the same model transport configuration using observations of beryllium-7 (7Be)".

Comment: L18-20, P25879: Is this result about summer exceedences being due to regional anthropogenic pollution discussed anywhere else in the paper? I know

the focus is on background ozone, but still it seems odd to have this conclusion here without presenting evidence to back it up. I'd expand this discussion, maybe even include a figure showing the correlation with anthropogenic CO.

Response: We now state in the text "the model still systematically underestimates the observed high-ozone events with O3 > 75 ppbv (0.4% of the data in spring, 0.7% in summer). From correlations with model tracers we find that these events in spring are associated with stratospheric intrusions, as discussed below, and in summer with regional anthropogenic pollution. For the observed summertime high-ozone events, the model shows elevated CO enhancements from North American anthropogenic emissions (42 $\pm$ 30 ppbv; differences in the CO concentration between the standard simulation and a sensitivity simulation with zero North American anthropogenic emissions), significantly higher than those for the ensemble of data in summer (21 $\pm$ 14 ppbv; p-value < 0.01 from the t-test)."

Comment: L20, P25880: I'd like to see more explanation of this discrepancy between GEOSChem and CMAQ. Why does GEOS-Chem show a larger lightning influence? How does your approach for lightning NOx emissions differ from that of Kaynak et al. (2008)? Do you have evidence that the GEOS-Chem result is more accurate than the CMAQ one?

Response: We now state in the text "Kaynak et al. (2008) found in the regional CMAQ model that lightning NOx emissions increases surface ozone by generally less than 2 ppbv, but their results focused on urban areas particularly in the eastern US."

Comment: L5-7, P25881: Have you examined whether the discrepancy at these sites could be due to a transport error in the model, so that the modeled smoke is hitting the wrong receptors? Do you have other tracers measured at or near these sites (like

CO, HCN, OC, etc.) that would show that wildfire smoke was present at the times the model predicted, but still there was no impact on surface O3? Also, what is the size of the excess O3 to excess CO ratio in the modeled smoke plumes (if available)?

Response: We added in Figure 8 two panels showing OC aerosol measurements from collocated IMPROVE sites. Elevated OC measurements confirmed wildfire impacts at the two sites. There is no CO measurement available for us to compute the O3/CO ratios in wildfire plumes.

We added in the text "The right panels of Figure 8 show measurements of organic carbon (OC) aerosol at collocated sites of the Interagency Monitoring of Protected Visual Environments (IMPROVE) (http://vista.cira.colostate.edu/improve/). We can see elevated OC measurements when the model simulates high wildfire ozone enhancements. However, the measurements show no correlated ozone enhancements that would indicate ozone production in the fire plumes."

Comment: L10-12, P25881: Why do you average the OC and O3 results over the domain? If fires are depleting O3 near the fire (through NO titration) and increasing it downwind, wouldn't your domain averaging simply wash away that signal? Also, the locations of the IMPROVE and CASTNET sites are not always the same, and IMPROVE has much larger coverage according to Fig. 4. Could this influence your results? What does the OC correlation look like if you only include the IMPROVE sites nearest to the CASTNet sites?

Response: We added in the paragraph "We further examine whether wildfire emissions would lead to regional enhancements of ozone concentrations.".

We also added in the text "CASTNet and IMPROVE sites have different spatial distributions (Figure 4), so we also examined the correlation for the subset of IMPROVE sites collocated with CASTNet. We find the same positive correlation for OC aerosol

as shown in Figure 9."

Comment: L2-5, P25883: I am confused at how Figure 10 relates to Figure 9. In Figure 9 we have no correlation between domain-average ozone and carbon burned in the (preceeding?) five days for the summers of 2006-2008. But Figure 10a shows that summer- and domain-averaged area burned and ozone are correlated with temperature between 1990-2008. If this is so, why is there no O3 to carbon burned correlation in Figure 9? Aren't these two results inconsistent?

Response: We add here in the text "The summer mean ozone enhancements over the Intermountain West are thus not directly associated with wildfire emissions, consistent with Figure 9 that shows no correlation at the daily time scale."

Comment: L10-11, P25885: You say the difference is "in part" due to the different definitions of stratospheric influence. Does that mean there is a significant difference remaining even when GEOS-Chem uses the Lin et al. (2012) approach? If so, how much of the difference remains?

Response: We now state "Lin et al. (2012) reported a higher stratospheric influence in their AM3 model simulations, 2–3 times greater than GEOS-Chem estimates for the western US. We see from the above that about half of the difference reflects a difference in definition of stratospheric influence, not an actual physical difference. The remaining difference reflects the role of vertical transport that is more vigorous in AM3 (Fiore et al., 2013).".

Add reference: A.M. Fiore, J.T. Oberman, M. Lin, L. Zhang, O.E. Clifton, D.J. Jacob, V. Naik, L.W. Horowitz, and J.P. Pinto: Estimating North American background ozone in U.S. surface air with two independent global models: Variability, uncertainties, and recommendations, Atmospheric Environment, submitted, 2013.

Comment: L15-16, P25887: You should also mention the lightning NOx yield changes due to moving the tropical/extratropical border here.

Response: We have shortened this sentence in the conclusion to avoid repetitive statements. It now states "We find that our improved lightning simulation largely corrects previous ozone overestimates by Zhang et al. (2011) over the Southwest US in summer." The improved lightning simulation has been discussed in details in the main text.

Comment: L25-27, P25887: I'd be more specific here, saying that the domain-average CASTNet ozone data showed no correlation with wildfires in the domain, in contrast to domain averaged OC from the IMPROVE network.

Response: We now state "Regional ozone concentrations averaged across the Intermountain West show no correlation with wildfires, in contrast to OC aerosol observations from IMPROVE sites that show strong correlation."

Typos and style Comments: L18, P25873: I would think background generally would mean the absence of any anthropogenic influences, not just local.

Response: We now state "in the absence of local or regional anthropogenic influences".

Comment: L5, P25874: The wording here is a little awkward. I'd suggest changing this to "Understanding the natural sources contributing to elevated ozone in the Intermountain West is of crucial importance for policy."

Response: Changed as suggested.

Comment: L 3, P25876: Expand "OTD/LIS"

Response: We now state "10-yr averaged satellite lightning observations from the Optical Transient Detector (OTD) and the Lightning Imaging Sensor (LIS)".

Comment: L25, P25876: I've generally seen "GFED2", not "GFED-2"

Response: Changed as suggested.

Comment: L24, P25880: Given that you conclude that fires had little noticeable impact on surface O3 during this period, I'd change this to "In the model, wildfires increase ozone by up to 20 ppbv" to make clear from the beginning that you think it is a model error rather than a correct estimate of the impact of fires.

Response: Changed as suggested.

Comment: L18, P25886: You don't need to redefine "CTM" here.

Response: Changed as suggested.

Comment: P25900, Fig 4: The caption implies that the sites discussed in the text should be labeled on the map, but I don't see the labels in my version.

Response: The figure is updated to include the labels.

Comment: P25903, Fig. 7: The title on panel (d), “Stratosphere (transported)” is misleading, as what is actually plotted is the Lin et al. (2012) definition of stratospheric influence which includes both chemical production and transport, if I understand it correctly.

Response: Yes, we now change the title of panel c) to "Stratosphere (GEOS-Chem definition)" and panel d) to "Stratosphere (e90 tracer)".

Comment: P25904, Fig 8: It’s hard to see the data points once all the other curves are plotted on top. Could you plot the black curve and points on top of all the others to emphasize the CASTNet data more?

Response: The figure is updated as suggested.

Comment: P25906, Fig. 10: The caption should mention that the bottom panel is only for 2006-2008.

Response: We now state in the caption that “The bottom panel shows spatial and interannual correlations for individual CASTNet sites for 2006-2008”.

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