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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Wildfire influences on the variability and trend of summer surface ozone in the mountainous western United States" *by* Xiao Lu et al.

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Comment: Overview: The paper presents a new approach to examining the influence of wildfire smoke on ozone mixing ratios at remote/rural monitoring sites in the U.S. intermountain west. Overall the paper is well written and suitable for publication in ACP. I recommend that the authors consider the following ideas in revising the manuscript. **Response:** We thank the reviewer for the valuable comments. All of them have been implemented in the revised manuscript. Please see our itemized responses below.

Comment: 1) Line 285: The sentence staring with "These underestimates" requires substantially more justification/analysis/references.

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Response: We move the original Figure S4 that shows these model underestimates to the main text (Figure 3), and now state:

"These underestimates, however, are not likely due to model underestimates of wildfire ozone influences. We show in Figure 3 the relationships of TFI values with measured MDA8 ozone, MLR wildfire ozone enhancements, and MLR residuals to assess the model performance for the subset of high ozone days (MDA8 > 70 ppbv). The MLR model residuals for those high ozone days have little correlation with TFI, and most of the model underestimates occur when there are small fire impacts or fires not captured by the FLEXPART retroplumes. We suggest that these underestimates may be associated with other factors not included in the statistical model such as transport from Asia or California, from lightning emissions or stratosphere. These processes could episodically produce more than 10 ppbv ozone in summer over the US Intermountain West (Zhang et al., 2014)."

Comment: 2) Line 315: There are many reasons that a model like GEOS-Chem will not adequately represent the role of fires. The standard versions of GEOS-Chem do not emit short lived VOCs, and the emission factors for NOx emissions from fires are quite variable in reality. The model also adds all the emissions within the boundary layer. The authors clearly recognize this because they use a 5km cut off for the FLEXPART analysis, and are certainly aware of recent work by Val Martin et al. (e.g. 2010) with respect to plume heights over North America. This should be discussed in depth or omitted. A reference to Zhang et al., (2014) is inadequate.
Response: We agree and add more text discussing why GEOS-Chem may not adequately represent wildfire chemistry.

We now state: "We can see that GEOS-Chem simulates up to 40 ppbv wildfire ozone enhancements for the short-distance sites, much higher than the MLR estimates (mean value of 3.96 ppbv versus 1.85 ppbv). A sensitivity simulation with a reduced wildfire NOx emission factor (from 3.0 g to 1.0 g NO per kg of dry mass burned)

would decrease the GEOS-Chem mean ozone enhancement for the short-distance sites from 3.96 ppbv to 2.06 ppbv. On the other hand, for the long-distance sites, the GEOS-Chem wildfire ozone enhancements become substantially lower than MLR (0.77 ppbv versus 1.02 ppbv). We see GEOS-Chem largely overestimates wildfire ozone influences near the source regions but fails to capture continued ozone production in wildfire plumes downwind, as also pointed out by Zhang et al. (2014). It reflects the difficulties for Eulerian models such as GEOS-Chem to simulate wildfire ozone production due to, e.g., missing short-lived VOCs (Jaffe and Wigder, 2012), inadequate PAN chemistry (Alvarado et al., 2010; Fischer et al., 2014), and limiting all fire emissions in the boundary layer without considering their injection heights up to the troposphere (Val Martin et al., 2010; Sofiev et al., 2013)."

Comment: 3) Why does this paper narrowly focus on the intermountain west? This region has many wildfires, but the smoke travels and the impact on ozone may be larger downwind (see Brey and Fischer, 2016). S. Brey and E.V. Fischer (2016), Smoke in the City: How often and where does smoke impact summertime ozone in the United States, Environ. Sci. Tech., DOI:10.1021/acs.est.5b05218.
Response: This study follows our previous work of Zhang et al. (2014), which focused on the Intermountain West where background ozone concentrations are high and the ozone trends are not fully understood as we described in the Introduction. It also demonstrates feasibility of our statistical approach to quantify wildfire ozone influences. We expect future work to apply the approach to other regions in the US or over the world.

We state in the Conclusion: "A recent study by Brey and Fischer (2016) investigated fire impacts on ozone at urban sites over the contiguous US, and found that fire ozone influences can be even higher at locations with high NOx emissions."

Comment: 4) I have two questions with respect to Figure 7 (and the associated discussion). First, is it appropriate to use the entire range of 1989-2010 to look at

the number of exceedance days. There have been trends in ozone during this time. Second, and more importantly, would it be more appropriate to view the exceedance days as a percentage of the total, rather than as a count. Yes, there will be more exceedance days as we lower (tighten) the ozone standard, all things held the same. However, do we have a way to determine if the relative importance of fires will increase?

Response: To answer the two questions: (1) Figure 7 (now Figure 9) has shown the time series of summer ozone exceedance days during 1989-2010. There is no significant trend in the exceedance days with or without wildfire impacts. And (2) we also calculate the percentage of wildfire contributed vs. total exceedance days, but find no increase in the relative importance of fires as lowering the ozone standard.

We now state in this section "We find no statistically significant trends in the number of exceedances for both the measured ozone concentrations and ozone in the absence of wildfires during the summers 1989-2010", and "In total, wildfires contribute 28%, 31% and 32% of the days with MDA8 ozone exceeds 65, 70, and 75 ppbv, respectively, reflecting small changes in the relative importance of wildfire influences as lowering the air quality standard over this region."

Comment: 5) Finally, I think all the SI materials should be moved into the main paper. There are very important figures in the SI materials, and I had to refer to them to follow the paper. Without them in the main manuscript, it would be easy to overlook the fact that the MLR really does not do a good job reproducing the highest ozone days. This is an important point in considering the value of this analysis.

Response: We now move the original Figure S3, Figure S4 (now Figure 2 and 3), and Table S1 (now Table 1) to the main text. Figure S3 and S4 explains that the MLR model underestimates of high ozone values are not associated with fire impacts. Tables S1, as also suggested by the Reviewer 2, shall be included in the main text. We think the rest can be kept as SI materials for limiting the length of the paper.

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