Responses to the Referee Comment #2 on acp-2021-485

The authors are addressing a much-needed topic in fire emission calculations and air quality modeling. When ground fuels burn they become a large source of trace gases and aerosols into the atmosphere and there is a great lack of data needed to quantify these emissions. This manuscript highlights the need for more information, and how the lack in the current available information hinders air quality analyses. Below I have specific comments designed to help make this analysis more robust.

Response: Thank you for reviewing our manuscript and providing very constructive and in-depth comments and suggestions.

Specific Comments

Fire Events: Four of the largest wildfire events in the southeastern US were selected for air quality modeling with WRF-Chem for two domains. One domain simulates fires in the southern Appalachian mountains and one domain simulates fires in southern Georgia. The fires ranged from 91K acres to > 500K acres. One thing I found lacking was consideration of the Evans Road and Pains Bay wildfires. They were smaller than the top 4 criteria (41K, and 5K acres respectively), but were significant in terms of the emissions from the burning of organic/ground fuels and subsequent air quality impacts. Rappold et al. 2011 conducted a health impact analysis from the Evans Road wildfire and Tinling et al. 2016 conducted a similar analysis for the Pains Bay wildfire. The first two sentences of the Abstract state that "Wildfires can significantly impact air quality and human health. However, little is known about how duff and peat burning contributes to these impacts." Given the goals of this paper, I would expect these studies/impacts be part of the introduction and also a consideration in this study.

Response: Thank you for this valuable comment. We paid attentions only to fire size when we were selecting fire cases for this study but ignored some important fires that occurred in the sites with rich organic soils despite relatively smaller size. As suggested, we conducted additional simulations of the 2008 Evans Road fire and the 2011 Pains Bay fire. The corresponding modifications to the manuscript are follows.

(1) Study region (Lines 166-169): We added a third area of coastal eastern North Carolina.
(2) Fire cases (Lines 192-199): we added simulation cases for the two fires in Table 1 with description in section 2.2. The 2008 Evans Road fire ignited on June 3, 2008 and burned 41060 Acres. The 2011 Pains Bay fire ignited on May 5 2011 and burned 29400 Acres. The simulation periods for the two fires were June 2008 and May 2011, respectively. A domain centered in North Carolina was used.
(3) Results: Two figures were added to illustrate the simulation results of the two fires. Figure 11 shows results from a typical fire day during the 2008 Evans Road fire (June 12, 2008), including the simulation-observation comparison in the Sim_nofire, Sim_FINN, and Sim_FINN_duff cases, the $PM_{2.5}$ enhancement due to duff burning, and time series from

June 7 to June 15 in an urban site and a rural site close to the burning region. Figure 12 is the corresponding results during the 2011 Pains Bay fire, including a typical fire day (May 12, 2011) analysis and the time series from May 6 to May 15. The evaluation of ozone is not shown because, similar to the 4 fire cases discussed in the manuscript, the ozone effect from duff is weak in comparison with the above-ground fuel effects. The results indicate that the PM2.5 enhancement from duff flaming is significant for both fire cases. The smoke transport effects more to the cities by the 2008 Evans Road fire and less during the 2011 Pains Bay fire, because the 2011 Pains Bay fire occurred under prevailing wind to the ocean.

(4) Discussion (Lines 455-461): We discussed the two cases in the revised manuscript,

Duff Flaming Phase Emissions: Why only focus on flaming emissions of duff? Smoldering phase emissions are important in terms of air quality impacts (as noted by Rappold et al. 2011 and Tinling et al. 2016). Smoldering of ground fuels does not always occur on a time scale of months to year, it can occur on the scale of hours/days, and while the plumes do not necessarily loft high, during the day they mix near the surface (where people breath) under the mixing height and can be transported further distances. At night they can transport along terrain features often impacting small towns in rural areas closer to the fires. This becomes an environmental justice issue as well. Limiting the work here to only flaming phase duff emissions unnecessarily limits the utility of this study.

Response: We agree with this point about the importance of smoldering smoke. Besides the two studies mentioned in the comment, recent studies such as Kim et al. (2018) and Chan et al. (2020) also emphasized the health effects of wildfire smoldering. We did not investigate smoldering smok for two reasons. First, smoldering smoke is mainly a local process. Most smoldering cases only impact towns and rural regions close to a burning site. This study focused on the regional air quality impacts of smoke, especially in remote large cities with dense populations. Secondly, WRF-chem we used is a regional model. The local smoke process during smoldering phase is not well described in a regional model. We are planning to dig into the duff smoldering phase more in a separate study using different modeling tools, such as the PB-P specific local smoke model (Liu et al. 2018). We added more discussion in the revised manuscript (Line 607-610)

Duff Consumption: Related to this is how much duff consumption actually went into each of the scenarios? I see that 4.6 cm of duff burned in the 2016 Rough Ridge fire which went into the App16 case (fuel loading 3.15 kg/m2). How many centimeters of duff burned in the Oke07, Oke11 and Oke17 cases? And was the same fuel loading from App16 (3.15 kg/m2) assumed for the Oke scenarios? The end of section 2.4.2 discusses how regrowth was handled, but again, what actual data went into the scenarios? I recommend adding the duff depth burned and fuel loading estimates to a table. Further, were all duff estimates assumed to burn in the flaming phase? Or were some estimated to burn in the smoldering phase (and thus eliminated if I am interpreting the discussion regarding the focus on flaming phase emissions)?

Response: The only measured data of duff burning used in this study was from the App16 case. Because of this limitation, we assume the 4.6 cm per day duff flaming rate for all the studied cases in the Southeastern US except Oke11 and Oke17. Duff at some grid points was already burned by

the fire prior to each of the two fires. We used a simple algorithm to estimate the depth of duff layer at the grid points. The lack of duff measurements in Oke sites and the two added sites is one of the biggest uncertainties for this study, which is discussed in the manuscript. We assess the uncertainty by conducting the sensitivity test which changes the duff flaming emission by ± 20% (discussed in Section 3.4). We clarified this in the revised manuscript (Lines 336-338) and added the duff depth burned and fuel loading in Table S4.

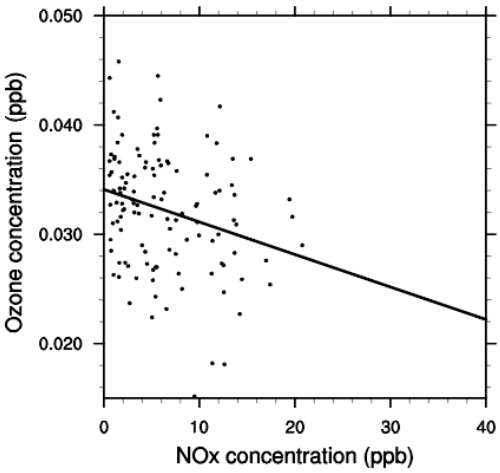
Yes, we only considered flaming burning in this study. The 4.6 cm duff burning is all from flaming phase. The carbon emission during the prolonged duff smoldering is larger in magnitude but with little regional air quality impact.

Duff Consumption: Section 2.4.2 indicates that "we estimated duff emissions and added them to FINN." Was a full suite of trace gas and aerosol species added? Or were only PM2. 5, NO and NO2 species added to the simulations? At a minimum I would expect a full suite of species using default above-ground fuel emission factors would be added to represent the duff fuels, and ideally those emission factors be adjusted based on available literature for duff fuels. Recent studies for the SE in George et al. 2016 and Black et al. 2016 may be useful. They both conducted lab experiments based on peat from North Carolina. Many of these trace gases have implications for ozone and secondary aerosol formation.

Response: Thanks for pointing it out. Not only NO_x and $PM_{2.5}$, we also had added a full suite of trace gas and aerosol species for duff emissions when we conducting the simulations. We included the duff emission of other species in the revised manuscript. In Table S2, we added the gas species duff burning emission factors used in this study. The duff emission species we added to the simulations are: CO, NO, NO_2, SO_2, NH_3, NMOC, BIGALK, BIGENE, $C_{10}H_{16}$, C_2H_4, C_2H_5OH, C_2H_6, C_3H_6, C_3H_8, CH_2O, CH_3CHO, CH_3COCH_3, CH_3COOH, CH_3OH, ISOP, MEK, MVK, TOLUENE, $PM_{2.5}$ (OC and BC). This list is based on the species list from the above-ground emissions in FINN v1.5. For the VOC species, the emission factors are from the organic soil burning emission factors summarized in Yokelson et al. (2013).

VOC-limited: Section 3.3. Is the SE (App16 domain) really in a VOC-limited scenario in the winter (Nov)? I recommend showing estimates to support this.

Response: The following estimations are used to confirm the VOC-limited scenario in the November of 2016. First, we show a relationship between the EPA measurements of daytime (8 am - 7 pm) ozone and NO_2 concentrations in Georgia and North Carolina. A negative correlation is shown in the figure below:



Then, in the response to the next issue, we made a sensitivity run that doubles the duff burning NO_x emissions. During the App16 case, increasing duff NO_x emissions further decreases ozone (Fig S26 in the revision), which also supports that November is in a VOC-limited scenario over the southeastern US. Some previous studies also mentioned that the ozone scenario is turning from NO_x-limited to VOC-limited from fall to winter in the eastern and southeastern US (Jacob et al., 1995; Simon et al., 2015; Zhang et al., 2016).

NOX and Ozone Generation: Much of the discussion focuses on how the duff did not add much ozone to the model simulations, which is attributed to the NO/NO2 emission factors being low. There needs to be more discussion about the variability in NO/NO2/NOX emissions from duff. Yokelson et al. 2013 is just a single experiment and Urbanski 2014 applies an uncertainty of 100% to the data. Studies such as Burling et al. 2010, McMeeking et al. 2009, Selimovic et al. 2018 and Clements and McMahon 1980 are all studies that measured emission factors for ground fuels. NO values range from 0.56 to 2 g/kg, and NO2 values range from 0.23 to 2.7 g/kg. These data argue for perhaps using greater EF values for NO and NO2 and also (especially) sensitivity runs that vary the NOX EF's by more than just 20%. Recommendation: I recommend an additional sensitivity run using 100% per Urbanski 2014 (e.g. 2x duff).

Response: One of the conclusions of this work is that when the PM effect of duff is as significant as that of the above-ground fuel, but the ozone effect from duff is not as strong as the effect from the above-ground fuel. For example, Burling et al. (2010) showed that the emission factor from duff is 0.738 g/kg for NO, and 0.232 g/kg for NO_2, and the emission factor from the above ground fuel is 1.720 ± 0.454 g/kg for NO and 1.023 ± 0.286 g/kg for NO_2 (Table 3 in Burling et al., (2010)). The summary of the NO_x emission factors mentioned in the reviewer's comment is included in a new table (Table S3). Corresponding description is added in the revised manuscript (Lines 514-520).

To address the reviewer's concern of the uncertainty due to the NO_x emission factor, we made sensitivity runs for the App16 and Oke07 cases using twice NO_x from duff burning (the '2x duff NOx' case). As shown in Fig. S26, doubling duff NO_x further decreased ozone during the App16 case. During the Oke07 case, ozone increases corresponding with the increased NO_x emissions from

duff, but the ozone effect is still weaker than the PM effect. The corresponding description is added in Section 3.4 (Lines 514-520).

Meteorology/Transport: Section 3.2 discusses the PM2.5 emissions and transport. I recommend making the discussion more robust by including references to support the statement "Both biases in fire emission calculation and smoke transport simulation should
be the contributors." I recommend Li et al. 2020 and Garcia-Menendez et al. 2013.

Response: Thanks for the suggestion. We cited Li et al. 2020 and Garcia-Menendez et al. 2013 in the revised manuscript.

Technical Corrections Table 1 caption needs to include more information such that the information in the table is understandable independent of the paper. Also, I recommend adding fire size (acres) to the table as well.

Response: As suggested, the title of Table 1 is revised to "The simulation period and fire emission inventories applied in different WRF-Chem simulations and experiments." Fire size information and the information about the new 2x_duff_NOx case is added in the table.

References used in the responses:

Jacob, D.J., Horowitz, L.W., Munger, J.W., Heikes, B.G., Dickerson, R.R., Artz, R.S. and Keene, W.C., 1995. Seasonal transition from NOx-to hydrocarbon-limited conditions for ozone production over the eastern United States in September. Journal of Geophysical Research: Atmospheres, 100(D5), pp.9315-9324.

Liu Y-Q, Goodrick S., Achtemeier G, 2018, The Weather Conditions for Desired Smoke Plumes at a FASMEE Burn Site. Atmosphere 2018, 9(7), 259; https://doi.org/10.3390/atmos9070259

Simon, H., Reff, A., Wells, B., Xing, J. and Frank, N., 2015. Ozone trends across the United States over a period of decreasing NOx and VOC emissions. Environmental science & technology, 49(1), pp.186-195.

Zhang, Y. and Wang, Y., 2016. Climate-driven ground-level ozone extreme in the fall over the Southeast United States. Proceedings of the National Academy of Sciences, 113(36), pp.10025-10030.