Responses to the Referee Comment #1 on acp-2021-485

The purpose of this study is to assess the importance of including duff in simulation of wildfire impacts on air quality. The authors conduct WRF-Chem simulations for four large wildfire events in the southeastern US using two fire emission scenarios – duff and no duff, as well as a control simulation with no fire emissions. The main findings of the study are:

1) relative to the no duff fire scenario (surface and understory fuels only), the increase in wildfire emissions through the inclusion of duff burning resulted in large increases in simulated surface PM2.5 concentrations near the fire locations (< 300km) and at remote urbans areas

2) while the no duff fire scenario increased regional O3 levels, the impact of additional emissions from duff burning were negligible for O3

3) relative to the control scenario (no fire) both the no-duff and duff fire simulations generally increased agreement between the WRF-Chem simulated PM2.5 and observations at surface air quality monitoring sites in fire impacted areas

4) relative to the no duff fire scenario, inclusion of duff emissions generally improved agreement of the WRF-Chem PM2.5 and observations at surface air quality monitoring sites in fire impacted areas

The authors conclude that modeling of regional air quality in the southeastern US can be improved by adding duff burning emissions to existing fire emission datasets and that emissions from duff burning have much garter impact on PM2.5 than O3.

The topic addressed in this study, contributions of duff and peat burning emissions to regional air quality, is certainly an important and of interest to air quality modelers and atmospheric scientists and is relevant to biomass burning in many regions. However, in the manuscript requires major revisions before it is suitable for publication. The paper is missing key methodological details and a couple important choices in the study design are not well justified. Additionally, the presentation and discussion of results and lacks definition and focus, making it difficult to evaluate the authors' conclusions and the overall broader relevance of the study.

Here I provide my most important concerns regarding the paper, followed by less crucial, specific comments.

Response: Thank you for reviewing our manuscript and providing critical and helpful comments. We revised the manuscript based on the comments to provide missed key methodological details and focus more on presentation and discussions.

1. Estimation of duff consumption by flaming combustion

In Zhao et al. (2019), post-fire field measurements at 4 sites (2 pair: 2 burned, 2 unburned) at the location of the ~11,700 ha Rough Ride Fire, indicated duff depth consumption of 4.5 cm and 4.9

cm. Based on undocumented and unreferenced information, "In fact, whereas a duff layer is typically consumed during the smouldering phase of combustion, the monitoring and images taken during the RRF indicated that a large portion of the duff layer burned during the flaming phase of combustion.", Zhao et al. (2019) assume that nearly all of the duff consumption occurred during flaming combustion in one day. In the current study, the authors use the 4.5 cm duff depth consumed by flaming consumption claimed by Zhao et al. (2019) and apply it to the four fire cases. This choice does not seem justified based on the less than robust information presented in Zhao et al. (2019).

Response: The photos below show night fire lines during the 2016 Rough Ridge Fire. Flaming is clearly seen. The duff layer was burned during flaming phase due to a half-year long severe drought prior to the fire. One coauthor of Zhao et al. (2019), Jeffrey Schardt served as a fire manager from the Chattahoochee-Oconee National Forests, USDA Forest Service monitored the entire development of the fire. We agree that there are no robust duff burning measurements in the Southeast US although a number of large fires have occurred in the duff-rich areas. Due to this limitation of direct measurements from different fire events, we had to apply the 4.5 cm duff burning depth to other fire cases. In Section 3.4, we discussed the uncertainty and conducted sensitivity runs to evaluate the corresponding variations of PM2.5 and ozone effects due to the duff emission uncertainty. On the other hand, this uncertainty was actually one of implications of the findings from this study for future fire emission and smoke air quality. The finding indicated the importance to measure and simulate duff burning for assessing the contributions of smoke to global air pollutions.

2. Duff PM2.5 emission factors

The study is simulating the impacts of flaming duff consumption on air quality, but they use PM2.5 EF factors for smoldering duff (Urbanski 2014; Geron & Hays, 2013). The high PM2.5 and VOC emission factors for duff burning result in large part because duff burns primarily by smoldering combustion. The authors should have used a reduced PM2.5 EF to represent flaming combustion.

Response: Thanks for this point. In Geron and Hays (2013), the peat and duff emission factors were measured simultaneously in lab experiments under simulated condition for peat and duff smoldering and flaming condition for other fuels (Section 3.2 in Geron and Hays 2013). There is limited information about the emission factor of duff flaming and its difference from duff smoldering, but individual and synthesis studies showed that the combustion efficiency of duff flaming is higher than smoldering (Lin et al. 2019), and that the flaming emission factor of $PM_{2.5}$ for some above-ground fuel is 30% lower than smoldering (Fig. 3 in Prichard et al. 2020). To further address the reviewer's concern, we conducted sensitivity runs tuning the duff $PM_{2.5}$ emissions by ± 30%, and discussed the potential bias and uncertainty due to duff emissions. The results of the sensitivity runs are shown in Figures S24 and S25. Despite the uncertainty from duff emission factors and fuel loading, tuning the duff $PM_{2.5}$ emissions by ± 30% did not affect the major conclusions.

In the revision, we included the uncertainty due to the different emission factors between smoldering and flaming (Line 352-354): "There were not enough duff measurements for the fire cases we investigated, and the duff emission factors between smoldering and flaming were also not well investigated.". Quantitative analysis in the Section 3.4 is also updated.

3. Temporal emissions profile

The authors do not describe how the daily fire emissions were converted into hourly emissions for the WRF-Chem simulation. The appear seems to imply they were not:
L408-409: "The daily variations are different between observations and simulations because the observed fire emission dataset was at daily rather than hourly intervals."

Response: The WRF-Chem simulations did not consider the diurnal cycle of fire emissions. FINN inventory analyzes the daily MODIS Thermal Anomalies Product and assumes the fire lasts for 1 day without diurnal cycle. Therefore, the fire emission applied in the WRF-Chem simulations for each hour is the hourly emission converted from the daily fire cases from FINN. We added the description into the revised manuscript (Line 256-258): "No a-priori diurnal cycle of the fire emission was applied in the WRF-Chem model, and the hourly fire emission applied in the WRF-Chem simulations was the hourly emission converted from the daily fire cases from FINN assuming each observed fire hotspots last for one day."

4. Assessment of smoke impacts

It is unclear how the authors define air quality (AQ) observation sites as influenced or not influenced by smoke. Is smoke "influenced" defined from the perspective of the model e.g., air quality monitoring sites that were impacted by a conserved smoke in the WRFChem simulation or PM2.5 or CO levels greater than non-fire simulation? Or is smoke influenced defined by AQ observation e.g., PM2.5 > some threshold. The criteria for smoke influenced needs to be clearly defined. And the rational for the criteria explained.

There are too many figures and the accompanying discussions are difficult to follow. I feel the study would be better served had the authors focused on a handful of days using air quality sites that were smoke impacted, from the simulations' perspective using a clear, well defined definition of smoke impacted (e.g., WRF-Chem conserved smoke tracer, CO
levels, etc.)

Response: Thanks for pointing out the missing definition of the smoke influence. We updated the definition in the Section 3.1 of the revised manuscript (Lines 360-364): "Here we define the fire influence based on the $PM_{2.5}$ impact from fire. If the $PM_{2.5}$ difference between sim_nofire and sim_FINN is less than 1 μg m^{-3} over a specific region (and time), then this region (and time) is not influenced by fire smoke. This value is near the low end of the thresholds often used to assess the smoke impacts (Munoz-Alpizar et al. 2017, Matz et al. 2020). "

Because we focused on regional smoke impacts on different sites and from different fire events, the logic of picking the studied time period was based on the intensity of the fire.

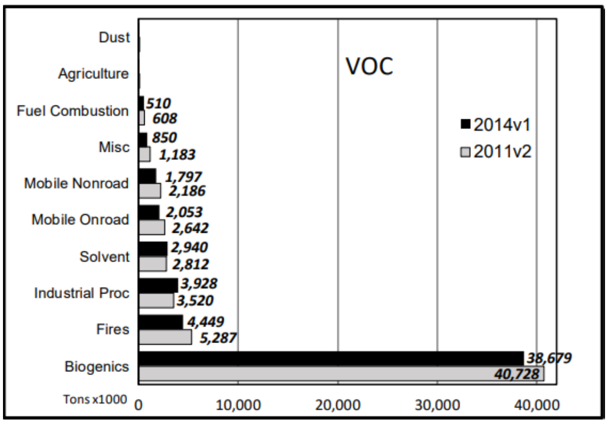
Specific Comments

5. L9: "The emissions of duff burning were estimated based on a field measurement"

Response: We rewrote the sentence as "The emissions of duff burning were calculated using the measured duff fuel consumption for the 2016 Rough Ridge Fire and emission factors."

6. L24-26: "Fires contribute 26.9% of total volatile organic compounds (VOC) emissions and 27.5% of PM emissions in the U.S. according to the 2014 US Environmental Protection Agency (EPA) National Emissions Inventory (NEI) (USEPA, 2017)."

Authors Response: Thanks for pointing it out. We double checked the EPA NEI 2014 report and rewrote this sentence as: "Fires contributed to 13.6% of total volatile organic compounds (VOC) emissions and 27.5% of PM emissions in the U.S. according to the 2014 US Environmental Protection Agency (EPA) National Emissions Inventory (NEI) (USEPA, 2017)." This number was calculated based on the following chart (a part of figure 4 of the NEI2014 report).



7. L33-34: "Wildfires produce about 3.5% of global tropospheric ozone production, though ozone production rates of individual fires vary with fuel type, combustion efficiency, etc. (Alvarado et al., 2010; Jaffe and Wigder, 2012)."

Location, time of year, meteorology, and pre-existing atmospheric composition are likely important factors as well.

Response: We modified the sentence to: "Wildfires produce about 3.5% of global tropospheric ozone production, though ozone production rates of individual fires vary with location, time, fuel type, combustion efficiency, meteorology, and local pre-existing atmospheric composition, etc."

8. L44-45: "In many regions around the world, including the U.S., wildfires have an increasing trend during recent decades…"
Elaborate on what kind of increasing trend? Frequency of large fires, fire severity, burned area?

Response: We modified the sentence to: "In many regions around the world, including the U.S., wildfires have an increasing trend during recent decades, in both the number and the area of total large fires (Dennison et al., 2014; Barbero et al., 2015). In addition, weather with high fire potential has appeared more frequently. (Yang et al., 2011; Jolly et al., 2015; Abatzoglou and Williams, 2016)…"

9. L49-55: Paragraph needs rewriting. Introductory sentence of the paragraph is about human health impacts of smoke, but two of three following sentences discuss radiative impacts of smoke aerosol. I suggest dropping the radiative impact sentences and added more information on health impacts.

Response: We dropped the radiative impacts sentences and filled more information on the health impact (Lines 50-56). the paragraph now becomes:

Negative impacts of wildfires on human health are devastating when fire plumes are transported to populated metropolitan areas (Kunzli et al., 2006). Epidemiological studies have revealed fire emissions' contribution to PM2.5 oxidative potential, which is related to respiratory and cardiovascular diseases (Verma et al., 2014; Yang et al., 2016; Fang et al., 2016). During the fire events in northwestern U.S. in August-September US, a regional mortality of 183 due to PM2.5 exposure was estimated, in which 95% was contributed by fire emissions (Zou et al., 2019). Based on the U.S. respiratory hospital admissions and additional premature deaths during and after fire events, the economic loss is $11 – 20 billion due to short term exposures, and $76 – 130 billion due to long-term exposures (Fann et al., 2018).

10. L71-72: "Duff typically represents the detritus or dead plant organic materials fallen at the top layer of soil."
This is a good location to define the terms "duff", "peat", and "organic soil". They are often used interchangeably when discussing global wildland fire. For example, a couple sentences down the authors use "organic soil".

Response: Responded together with the next comment.

11. L82-83: "Besides duff, peat is another burnable organic soil that typically represents the fermentation below the duff layer (Frandsen, 1987)."
See previous comment.

Response: We moved the sentence in L82-83 "Besides duff, peat is another burnable organic soil that typically represents the fermentation below the duff layer (Frandsen, 1987)" back to L72-73, and added another sentence after this: "'Organic soil' is often used to represent soil formed by plant and animal decomposition, including peat and duff. Duff, peat and organic soil were sometimes used interchangeably, and we focus on duff in this study."

12. L105: Define "prescribed fire"

Response: We changed this sentence to "On one hand, most fires in the southeastern US are prescribed (planned) and…" (Line 107-108).

13. L106: Yokelson et al. (2013) is not a good reference for this statement, more appropriate reference(s) needed.

Response: We changed the reference to the following report from US Forest Service:

Waldrop, T.A. and Goodrick, S.L., 2012. Introduction to prescribed fires in Southern ecosystems. Science Update SRS-054. Asheville, NC: US Department of Agriculture Forest Service, Southern Research Station. 80 p., 54, pp.1-80.

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