Reviewer #1

We kindly thank the reviewer for his/her time to evaluate our manuscript, and we appreciate the positive view on the work presented. Our comments appear below in red.

Specific comments:

The authors designed four synthetic WRF-Chem experiments driven by different biomass burning scenarios representative of different regions but all the simulations were performed over South America. Since the chemistry and weather of boreal forests, Africa, and Australia are different than South America, the simulated NO2 and CO columns could have been different for boreal forests, Africa, Indonesian, and Australian fires had the WRF-Chem domains been set-up over each region representing a different biomass burning characteristics because meteorology and chemistry over each biomass burning region is different from South America. I suggest the authors to include a discussion on this aspect.

To clarify, we did WRF-CHEM experiments first and foremost to demonstrate the effectiveness of the Statistical Bulk Method (SBM) of sampling CO and NO_2 data relative to a background. In these experiments we used by design the same meteorological and chemical conditions to focus mainly on our ability to retrieve different types of burns using SBM technique. We do acknowledge that variability in meteorology and chemistry can play a significant role to CO, and in particular, NO_2 concentrations. For instance, additional sources of CO can come from oxidation of volatile organic compounds (VOC) and methane and removal of CO can come from oxidation with hydroxyl (OH) radicals. Dekker et al. (2019) found that these combined processes had in the end very little impact on CO concentrations. The chemistry of NOx is complex as well, as the cycling between NO and NO_2 takes place within minutes to hours. During the daytime there is a photochemical balance between NO_2 photolysis and NO oxidation by ozone converting NO into NO_2. The principal sink of NOx is the oxidation to HNO_3 in reaction with OH.

To better investigate whether how meteorology and different chemistry regimes would impact CO and NO_2 we sampled other regions within the South American WRF domain. In Fig. 1 here below we show for each type of burn the daily and monthly average mole density ratio (MDR) between NO_2 and CO sampled from WRF-CHEM and TROPOMI. For each burn we show four different WRF-CHEM MDR estimates:

(1) standard simulation with sampling at the default region (as in the paper),

(2) simulation with sampling at region 1 with every day a repeat of the same meteorology,

(3) standard simulation with sampling at alternative region 2,

(4) simulation with sampling at alternative region 2 with every day a repeat of the same meteorology.

The different chemistry regimes are mimicked by sampling in an alternative region more east in the WRF-CHEM domain, closer to the Atlantic Ocean. Other meteorological regimes are mimicked by applying constant meteorology as an alternative simulation.

As shown in Fig. 1, both meteorology and chemistry do play a role in affecting the MDR estimate. For burns where NO_2 emissions are relatively more prevalent, like for deforestation and savanna fires, the differences are naturally exacerbated. NO_2 is more affected by the chemical conditions and meteorology than CO. This is expected and is also observed in the TROPOMI data in Fig. 6a of the new manuscript (Fig. 11a of the original manuscript), where we have quite some variability in MDR for the savanna and deforestation fires across the different regions. Across the ensemble of four simulations we do find on average the same MDR signatures as detected by TROPOMI. To accommodate the concern of the reviewer, we acknowledge this in the text in Discussion Section 4, starting at line number 709: *"In general, a large part of the biases in ΔXNO_2 (and thus in MDR), either caused by the sampling techniques or the instrument precision and sensitivity, were in all likelihood somewhat similar in magnitude in the regions we studied. Hence, we believe it did not impair the detection of differences in fire characteristics. The uncertainty related to chemistry and transport may have played a larger role region-to-region as it affected tropospheric NO_2 more differently than CO, and thus our ability to derive a robust MDR. In particular, on shorter day-to-day time scales the MDR estimates can vary greatly. The amount of OH radicals in the atmosphere acts as the primary daytime sink of NO_2 and can vary substantially depending on the amount of tropospheric O_3, water vapor and incoming sunlight (source of OH), and the presence of other chemical species such as volatile organic compounds (sink of OH). Overall, it reduces the lifetime of NO_2 to several hours, much shorter than the lifetime of CO. As a consequence, daily estimates of ΔXNO_2 will always be biased low. In addition, daily variations in ΔXNO_2 that are driven by transport and chemistry are naturally exacerbated in $\Delta XNO_2/\Delta XCO$ ratio-space. Therefore, to interpret MDR, it is currently necessary to collect multiple days of data (e.g. for an entire month) to retrieve a more robust combustion efficiency signature that cancels out some of the day-to-day variations in transport and chemistry."*

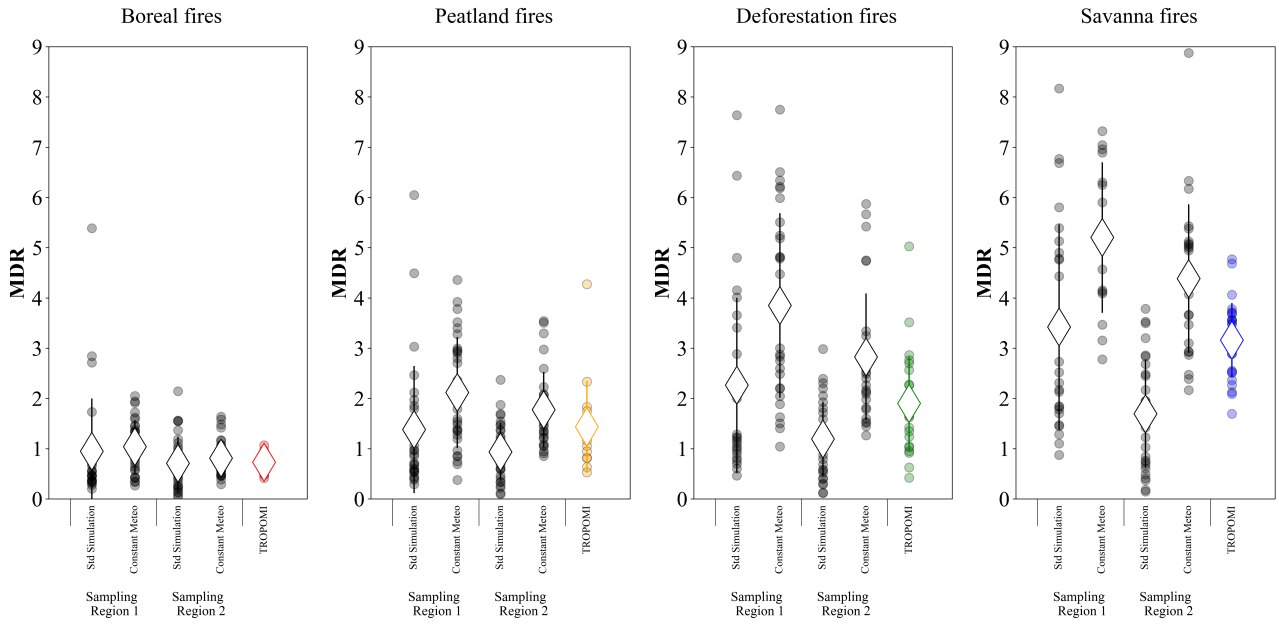

Figure 1. The daily mole density ratio (MDR) between NO_2 and CO derived for four different types of burns; boreal fires, peatland fires, deforestation fires and savanna fires. For each burn four different WRF-CHEM experiments were performed outlined along the x-axis: (1) standard simulation as described in Sect 2.4 of the manuscript, (2) simulation with constant daily meteorology, (3) standard simulation sampled at the alternative region more east of the domain, (4) simulation with constant daily meteorology at the alternative region. The actual TROPOMI observed MDR based on real fires is shown by the colored symbols. The monthly average with 1σ standard deviations is shown by the diamond symbols.

Dekker, I. N., Houweling, S., Pandey, S., Krol, M., Röckmann, T., Borsdorff, T., Landgraf, J., and Aben, I.: What caused the extreme CO concentrations during the 2017 high-pollution episode in India?, Atmos. Chem. Phys., 19, 3433–3445, https://doi.org/10.5194/acp-19-3433-2019, 2019.

While it was interesting to learn about TROPOMI's ability to distinguish between different biomass burning characteristics, I felt the paper should also have included a discussion on the crop-residue burning. Is it difficult to perform a similar analysis for crop-residue burning (e.g., in China or northern India) because of the limited sensitivity of TROPOMI NO2 retrievals to PBL?

We agree with the reviewer that it would have been good to include agricultural burning. In fact, we have looked into this and had hoped to include results in the manuscript, but we chose not to include this type of burning in the current study for two main reasons:

1) When focusing on large-scale agricultural regions such as in Indo-Gangetic Plain (IGP) in India we indeed observed a strong buildup of CO and NO_2 during the pre-monsoon wheat burning season (April-May) and post-monsoon rice burning season (October-November). However, a recent study by Dekker et al. (2019) showed that residential and commercial combustion in the same area was a much larger source of pollution than crop burning. The pollution was not only limited to New Delhi, but the accumulation of pollution extended over the entire IGP due to the meteorological conditions. This makes the differentiation between combustion of crops and other anthropogenic sources more difficult and so the ratio between NO_2 and CO is more likely to be misinterpreted.
2) The burning of crops is more difficult to interpret from space because the burning practices can vary quite substantially between farmers. A recent survey study by Liu et al. (under review) showed that there is a variety of burning practices and methods used in IGP (and other regions), resulting in either complete or partial burning of the crop residue. This is in particular important because it affects trace gas measurements from these burns in several ways. For instance, partial burns are less likely to be observed from space than complete burns. Partial burns also tend to release more particular matter and CO than complete burns due to smoldering combustion of wetter residues.

We therefore feel a dedicated paper focusing on the various forms of agricultural waste burning with a more regional focus would more informative and better justify the complexity of this fire type.

Liu, T., Mickley, L.J., Singh, S., Jain, M., DeFries, R.S., and Marlier, M.E.: Crop residue burning practices across north India inferred from household survey data: bridging gaps in satellite observations, EarthArXiv, under review, 2020.

Small Comments:

Section 2.4: Are the fire emissions subjected to plume rise in WRF-Chem?

We did not use the intrinsic plume rise calculations from WRF-CHEM. Instead, we applied a gridded characterization of measured injection height profiles for South America based on remotely sensed stereo-height information from smoke plumes (Martin et al., 2018). This allowed the trace gases from fires being emitted more realistically at multiple vertical levels in the planetary boundary layer instead just at the surface. This information was omitted by mistake in the original manuscript. In the new manuscript we explicitly mention this. In Section 2.4, at line number 403, page 20 we now write *"In addition, we used a spatial characterization of injection height profiles based on space-based stereo-height information from smoke plumes (Martin et al., 2018)".*

Martin, M. V., Kahn, R. A., and Tosca, M. G.: A global analysis of wildfire smoke injection heights derived from space-based multi-angle imaging, Remote Sens., 10, 1609; doi:10.3390/rs10101609, 2018.

Line 432: Based on legends of Figure 5, I think 2.77 should be replaced with 2.97.

Correct, we changed that in the new manuscript

Line 508: Change "EF in GFED4s" to "EF in neither GFED4s"

Changed in the new manuscript

Line 553: remove "the" before "efficient".

Changed in the new manuscript