

Reviewer #2

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.

Despite many information provided and careful investigation, it needs to be shortened to focus on the main findings. The lengthy descriptions of each session can distract the main points of the originality of the work.

We have realized that our original manuscript has been lengthy. We appreciate the arguments made by the reviewer to focus on the main results. We have carefully searched for redundancies and we have brought the important results more to the foreground. More specifically, the results Section 3.1 now starts with the global signatures in  $\text{NO}_2/\text{CO}$  (former Fig. 11, now Fig. 6) and we compare those with the simulated ratios from WRF-CHEM simulations. Subsequently, we show how the signatures compare with the efficiency signatures imbedded in the fire inventory products, GFED4s and GFAS (former Fig 12, now Fig. 7). We also include a new Table 2, showing all the values of MDR and EFR for the different regions (see specific point below). We left out the somewhat redundant discussion of the TROPOMI signatures for the different subregions (former Section 3.2). The main results and discussion of these regions are already included in Section 3.1 and Fig. 6. In Section 3.2 (former Section 3.1), we discuss the results of the South American deforestation and savanna fires in more detail, and the validation of the different sampling techniques. All in all, this reduces the text with 1200 words, we lose one figure (original Fig. 9), and we feel this improves the flow of the paper

#### Specific comments

The ratio between  $\text{XNO}_2$  and  $\text{XCO}$  implies not only the information on the surface emissions but also the information of its transport especially considering the longer lifetime of  $\text{CO}$ . How do you think the column comparison can cause the uncertainties of surface emissions? The author may have to comment on this concisely.

We agree that uncertainties arise from atmospheric transport and differences in lifetime of  $\text{CO}$  and  $\text{NO}_2$ . We discuss this in the last paragraph of the Discussion Section 4 in the new manuscript on page 34, starting at line number 709: *“In general, a large part of the biases in  $\Delta\text{XNO}_2$  (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  $\text{NO}_2$  more differently than  $\text{CO}$ , 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  $\text{OH}$  radicals in the atmosphere acts as the primary daytime sink of  $\text{NO}_2$  and can vary substantially depending on the amount of tropospheric  $\text{O}_3$ , water vapor and incoming sunlight (source of  $\text{OH}$ ), and the presence of other chemical species such as volatile organic compounds (sink of  $\text{OH}$ ). Overall, it reduces the lifetime of  $\text{NO}_2$  to several hours, much shorter than the lifetime of  $\text{CO}$ . As a consequence, daily estimates of  $\Delta\text{XNO}_2$  will always be biased low. In addition, daily variations in  $\Delta\text{XNO}_2$  that are driven by transport and chemistry are naturally exacerbated in  $\Delta\text{XNO}_2/\Delta\text{XCO}$  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.”*

Basically, we argue that it is necessary to collect data for an entire month opposed to a single day to retrieve more robust combustion efficiency signals. Potentially, we could translate the TROPOMI retrieved column enhancement ratios into emission ratios by accounting for NO<sub>2</sub> removal by OH similar to what is demonstrated in Lama et al. (2019). This is something we would like to explore in the future; however, it requires more detailed separate analysis for each region. This study focuses on finding differences in combustion efficiency on a global scale using TROPOMI NO<sub>2</sub> and CO data “as is” without applying additional correction from other sources.

*Lama, S., Houweling, S., Boersma, K. F., Aben, I., van der Gon, H. A. C. D., Krol, M. C., Dolman, A. J., Borsdorff, T., and Lorente, A.: Quantifying burning efficiency in Megacities using NO<sub>2</sub>/CO ratio from the Tropospheric Monitoring Instrument (TROPOMI), Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2019-1112>, in review, 2019.*

Are the types of burning affected by the soil and its condition? I wonder if more factors impact on the burning conditions (XNO<sub>2</sub> and XCO ratios).

Environmental conditions, like soil moisture, do indeed play a role in affecting the combustion efficiency. For instance, combustion of wet organic matter tends to be less complete producing relatively more CO than NO<sub>2</sub> and CO<sub>2</sub>. This is probably one of the reasons why we see quite some day-to-day variability in sampled  $\Delta$ NO<sub>2</sub> and  $\Delta$ CO and their ratio in Fig. 6a of the new manuscript (old Fig. 11a). We can only distinguish deforestation fires from savanna fires at the peak of the Amazonian fire season, in September. The monthly average mole density ratio (MDR) is then significantly different for the two fire types. The emission factors for NO<sub>2</sub> and CO that are used by the fire emission inventories are based on a large number of different field and laboratory experiments. The average EF values that are listed in Table 1 of the manuscript do not necessarily reflect these natural variations of burning conditions. In the Introduction, at line number 70, we make the argument that the actual EFs could be different from the biome averaged values used by the fire models. Variations in the chemical and structural composition of biomass, temperature, moisture content, and wind speed can all affect combustion efficiency. Our paper aimed to understand the broad variability and we necessarily averaged over larger regions and longer timescales (see previous comment) to average out some of the variability that may be introduced on finer scales but which requires more careful accounting for transport and chemistry, amongst others.

I think the information in Figure 11 and 12 is better in the table. The table of EFs would be useful with the regions, types of burning, and seasons. That would be useful for scientific communities.

We thank the reviewer for this suggestion. We removed the barplot that is part of the old Fig. 12 (now Fig. 7) because it adds no new information and instead we inserted Table 2, which provides an overview of all MDR and EFR values derived from TROPOMI data and WRF-CHEM.

Are the ratios of deforestation fires different from all types of vegetation fires? Then it means we can capture the deforestation by MDR from space? Please describe the meaning of identifying the deforestation by the satellite sensing.

From the burns investigated, we do find that the bulk of deforestation fires in South America are clearly less efficient than savanna fires, but more efficient than peat and boreal fires. It does indicate a greater contribution from smoldering combustion of organic soils and woody debris that is typically piled together at the surface. While promising, the regional burning characteristics are currently detected at a limited length scale of 500-1000 km and at a time scale of a month. The day-to-day variability in MDR remains quite large due to natural variations of individual burns and variations in meteorology and chemistry to accurately retrieve daily combustion efficiency signals. That is why it is currently not possible with this method to pinpoint deforestation from illegal logging or mining activities at a more local level.