Title: Direct estimates of biomass burning NOx emissions and lifetime using daily observations from TROPOMI
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General comments:

This manuscript show the capabilities of NO2 satellite retrievals from the TROPOspheric Monitoring Instrument from wildfire smoke plumes. An exponentially modified Gaussian (EMG) approach, NO2 TROPOMI retrievals, MODIS FRP, aerosol layer heights from TROPOMI, and reanalysis data are used to estimate the fire emissions of NO2 and its lifetime in the smoke plumes from >3000 fires globally. Fire locations and intensity are derived using MODIS Fire Radiative Power (FRP). The authors used GEOS-CF to modify the NO2 a-priori profile to correct the low bias of NO2 retrievals over fire plumes. The authors made a detailed comparison of their results with previous references regarding biomass burning emissions. Finally, the authors found that there is an anticorrelation between fire size and NO2 lifetime, possibly attributed to the higher emissions of HOx in larger fires. The manuscript has detailed methods, has an in-depth discussion of uncertainties, results are interesting and presented in a clear way, and is well written. Therefore, I suggest publication after minor revisions.

Specific comments
L66: looks like a good place to introduce PAN and organic nitrates?
L83 and elsewhere: It would be good to specify the local time of TROPOMI overpass. I am guessing that this influenced the choice of the NOx/NO2 ratio. I would like a little more discussion about how this ratio can change as time of the day. Is there any variation of it as the plume ages?
L153. I had a little bit of trouble understanding if the calculated lifetime included dilution. Reading the manuscript a second time, it was clearer. Is there a way that you can separate dilution from chemistry (i.e., using a NO2 normalized excess mixing ratio with respect to CO or CO2?). This might help you to further constrain or interpret your idealized plume model.
L175 Repeated comment but I would like to see more discussion about NOx/NO2 ratio as a function of the time of the day or distance from plume. References that might be helpful include Yokelson et al., 2009; Akagi et al 2011; Alvarado et al 2010; Juncosa Calahorrano 2021).
L195 Section 3.4 Did you used a plume specific NO2 background (i.e., for the background condition the day the plume was retrieved by TROPOMI)? For locations with fire seasons that last for months, background conditions can change because of the presence of dilute smoke in the area.
L220. Please include more details on figure S1 e.g., rotation angle. Might be good to have a final figure after rotation as well
L221 Can you please explain how you differentiate between the fire center vs. the apparent fire center? Is the first based on the FRP and the latter based on visual inspection? Also, this line needs more detail. What do you mean by “give good fitting statistics”? What is the correlation that needs to satisfy $R^2>0.5$?
Figure 1. I would have expected more tropical fires detected by TROPOMI. Did one of the criteria to remove plumes excluded those? Why? Also, it looks that there are not many fires in the equatorial line in South America and Africa, which is odd. Can you explain please? Perhaps this is an issue with the satellite retrievals?

L235 How did you identify Ag fires?
L247 and Figure 1 Very nice section. I was just wondering why you didn't remove the small fire towards the upper left side of the bigger fire. I assumed your criteria will remove it because it is clearly overlapping with the bigger one. Please explain.
L267 This sentence is confusing. I would remove the first part (before the comma) from this sentence.
L284 I am a little concerned about the conclusion that emissions and Fire Radiative Power correlate. In past field campaign, its being shown that reduced vs. oxidized emissions of nitrogen correlate very well with the Modify Combustion Efficiency (MCE) (i.e., smoldering vs. flaming) but it has been difficult to correlate them directly to FRP. The fire condition can also impact the chemistry in PECANS. If the fire has lower MCEs, there will be a lot of reduced nitrogen (e.g., NH3) compared to oxidized nitrogen (e.g., HONO that produces OH, NOx, etc). I know that getting MCE from many different fires is challenging, but I am not convinced that using FRP is the right approach. At the very least, there should be more discussion about how MCE affects emissions in the manuscript.
L333 Here is where I realized that the lifetime included dilution. Please include a few sentences somewhere earlier in the manuscript identifying all the loss processes that the estimated NO2 lifetime includes.
L465 I think you should discuss the thermal dependency of PAN. If the plume is injected high enough, PAN can be stable and thus its transportation can be very efficient (e.g., not a source of NOx close to the plume, at least in the time scales this relevant to this manuscript).
L468 We know well that PAN forms rapidly in fire smoke plumes and that its production plateaus after ~4 after of plume aging (Yokelson et al., 2009, Akagi et al., 2011, Alvarado et al., 2010, Juncosa Calahorrao 2021). It might be helpful to look at how the ratio of PAN/NOx changes as the plume ages and have some discussion about it.
L470 can you find another word for "estimate" so it is not right before "overestimate"
Great paper!

References

Yokelson, R. J., Crounse, J. D., DeCarlo, P. F., Karl, T., Urbanski, S., Atlas, E., Campos, T., Shinozuka, Y., Kapustin, V., Clarke, A. D., Weinheimer, A., Knapp, D. J., Montzka, D. D., Holloway, J., Weibring, P., Flocke, F., Zheng, W., Toohey, D., Wennberg, P. O., … Shetter, R. (2009). Emissions from biomass burning in the Yucatan. *Atmos. Chem. Phys.*, 28.

Akagi, S. K., Craven, J. S., Taylor, J. W., McMeeking, G. R., Yokelson, R. J., Burling, I. R., Urbanski, S. P., Wold, C. E., Seinfeld, J. H., Coe, H., Alvarado, M. J., & Weise, D. R. (2012). Evolution of trace gases and particles emitted by a chaparral fire in California. *Atmospheric Chemistry and Physics*, *12*(3), 1397–1421. https://doi.org/10.5194/acp-12-1397-2012

Alvarado, M. J., Logan, J. A., Mao, J., Apel, E., Riemer, D., Blake, D., Cohen, R. C., Min, K.-E., Perring, A. E., Browne, E. C., Wooldridge, P. J., Diskin, G. S., Sachse, G. W., Fuelberg, H., Sessions, W. R., Harrigan, D. L., Huey, G., Liao, J., Case-Hanks, A., … Le Sager, P. (2010). Nitrogen oxides and PAN in plumes from boreal fires during ARCTAS-B and their impact on ozone: An integrated analysis of aircraft and satellite observations. *Atmospheric Chemistry and Physics*, *10*(20), 9739–9760. https://doi.org/10.5194/acp-10-9739-2010

Juncosa Calahorrano, J. F., Lindaas, J., O'Dell, K., Palm, B. B., Peng, Q., Flocke, F., Pollack, I. B., Garofalo, L. A., Farmer, D. K., Pierce, J. R., Collett, J. L., Weinheimer, A., Campos, T., Hornbrook, R. S., Hall, S. R., Ullmann, K., Pothier, M. A., Apel, E. C., Permar, W., … Fischer,

E. V. (2021). Daytime Oxidized Reactive Nitrogen Partitioning in Western U.S. Wildfire Smoke Plumes. *Journal of Geophysical Research: Atmospheres*, *126*(4). https://doi.org/10.1029/2020JD033484