Reply to Reviewer 3

The authors directly estimate NOX emissions and lifetime for fires by using an exponentially modified Gaussian analysis of tropospheric NO2 columns observed by TROPOMI. The authors firstly correct the low bias of TROPOMI retrieved NO2 columns by replacing the a priori profile of NO2 with the GEOS-CF simulated profile at a finer resolution of 0.25. Representative NOx emission factors for six fuel types are derived by using the observations of fire radiative power from MODIS. The authors also discussed the uncertainties and capabilities of the method thoroughly. The scope fits ACP and the scientific idea is new. I recommend the paper be published after the authors address the following comments.

Reply: We would like to thank the reviewer for their constructive feedback and time spent reviewing this paper. Below is our response to reviewer's comments.

Major comments.

1. A better result is expected after the authors used a priori NO2 profile with 0.25 to replace the one used by the operational product. However, this spatial resolution is still much coarser than TROPOMI's, which implies that nearby pixels use the same profile shape. As the authors presented in the paper that fire events normally take place locally. How much uncertainty can it contribute?

Reply: We group fire pixels whose distances are within 20 km as a single fire event (~ 10 km radius). For most fire plumes we analyzed, the extent of the fire plumes is larger than the resolution of GEOS-CF (0.25˚). We agree that such resolution may not be able to resolve the spatial variation of NO_x within fire plumes, but the focus here is to derive an overall emission and lifetime estimate for the entire plume. As a result, we expect the *a priori* is largely converged with respect to spatial resolution.

We have added the following discussions in the revised manuscript:

The resolution of current global model simulation, however, is not sufficient to resolve the fine-scale chemical evolution of fire plumes, and better treatment of the fire injection is needed (Paugam et al., 2016). Assessment of the satellite retrieval uncertainty will benefit from high-resolution regional simulations combined with *in situ* measurements that sample individual fire smokes from the point of emission to downwind regions (Juncosa Clahorrano et al., 2021; Lindaas et al., 2020).

2. This plume-based method works only when the wind speed is not small, that is the plume exists. The authors keep every case even with very low wind speeds (< 2m/s). Should these cases be removed for the scientific reason?

Reply: We did not apply filtering for wind speed because the definition of 'calm' condition may be subjective. We assume EMG function should be able to identify the suitable cases as long as a satisfying fitting is achieved. There are indeed very few fire cases selected for calm winds (< 5%). To avoid confusion, we have removed the fires with wind speed less than 2m/s in Figure 5:

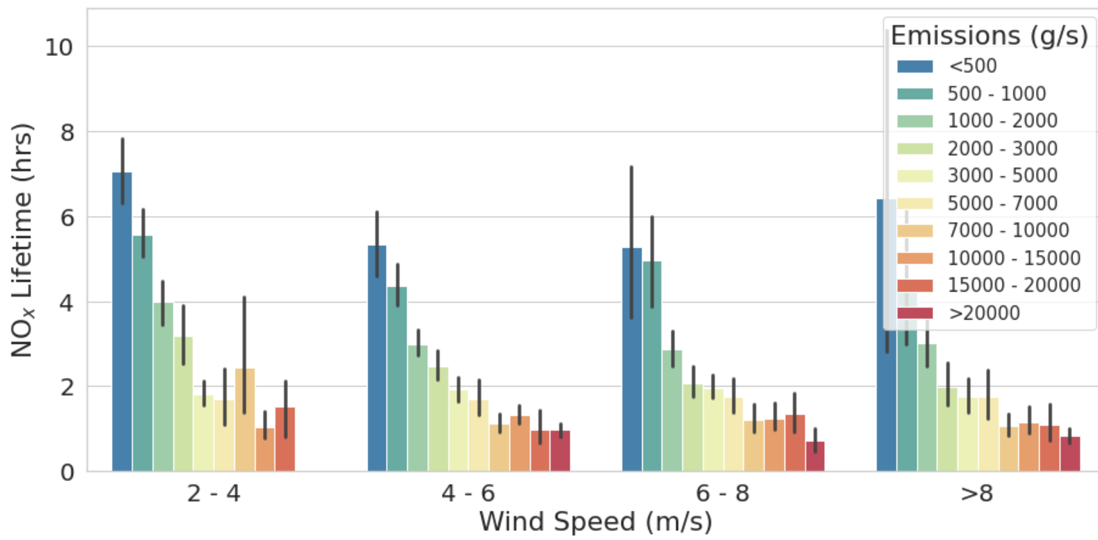

Figure 5 The mean and standard deviation of TROPOMI derived NO_x lifetime from fires at different emissions (colour) and wind speeds. Fire episodes with less than 2 m/s wind speed are not shown.

3. The authors argue that the difference between TROPOMI and OMI derived is mainly due to the a priori NO2 profiles, which is not accurate. The VCDs of TROPOMI are found to be lower than OMI's over many places, which is mainly caused by impropriate surface albedos or cloud pressures. Please give more solid proof if the authors want to draw this conclusion (i.e. section 4.4).

Reply: We agree that the *a priori* is not the only reason for the difference between TROPOMI and OMI. As we did not use OMI observations in this manuscript, the direct comparison between TROPOMI and OMI retrievals is beyond the scope of this study. We have revised the section to avoid confusion:

Using the standard TROPOMI NO_2 products without updating the *a priori* profile, the derived NO_x EFs are 44 to 66% of EF_{sat}, and 26 to 68% of $EFs_{andreae}$. Assessment of TROPOMI NO_2 with *in situ* measurements also suggest TROPOMI NO_2 is biased low over polluted regions, and replacing the coarse-resolution *a priori* profile with fine-resolution simulations could largely reduce the low biases (Judd et al., 2020; Tack et al., 2021). Our derived NO_x EFs are nearly 3 times larger than a previous study based on OMI observations, which suggest NO_x EFs are lower than 1g/kg in all fuel types (Mebust and Cohen, 2014). Besides the differences in satellite instruments and methods, the discrepancy is partially due to less accurate representation of biomass burning emissions in the *a priori* profile of NO_2 in Mebust and Cohen (2014). Using the standard TROPOMI NO_2 products without updating the *a priori* profile, the derived NO_x EFs are similar to those developed by Mebust and Cohen (2014) for boreal and temperate forest fires, but still higher over other fuel types.

4. The section 3.4 is long and complicated. A flowchart is helpful to explain the procedure or moving this part to the supplementary.

Reply: We have added a flowchart in the Supplement to explain the procedure of fire selection:

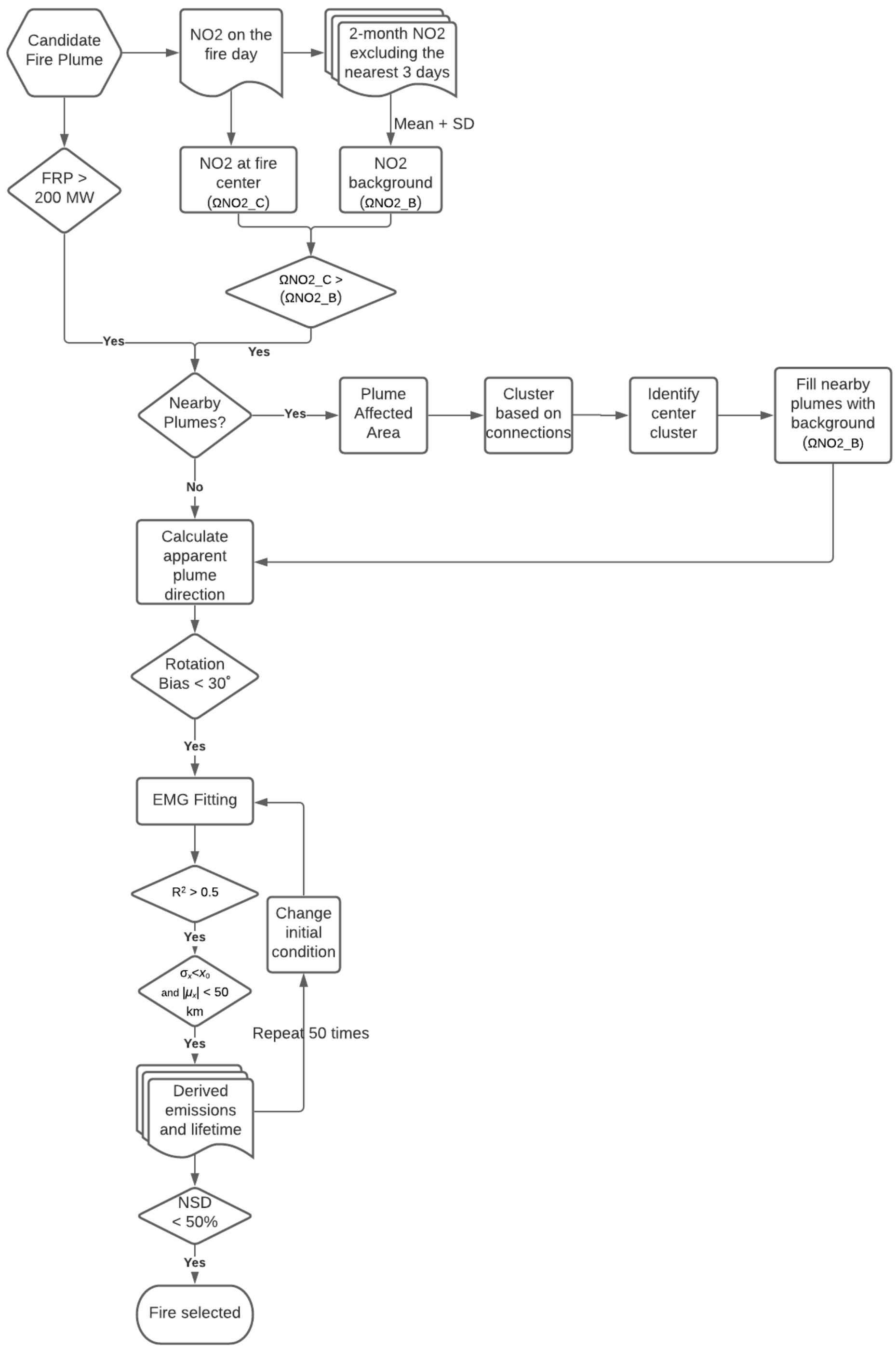

Figure S1 Flowchart that illustrates the processes to select candidate fires.

5. The authours intend to derive representative NOX emission factors for six fuel types. However, satellite observations are available once per day, and some fire events can last for several days. In these cases, the fire intensity and the chemical condition also change. The authours, at least, should give an example to explain how to consider the emission factor for a certain fuel type.

Reply: The fuel type classification is based on annual MODIS land cover type product, which is a function of location only, and should not vary temporally. We have clarified this point in the revised manuscript:

The fire episodes are classified based on MODIS detected fire location following the fuel classification in the Global Fire Emission Database (GFED), which is estimated using the MODIS land cover type product and University of Maryland classification scheme (Friedl et al., 2010; van der Werf et al., 2017).

We agree that fire intensity and the chemical condition may change, which cannot be captured by the single overpass of TROPOMI. We derive NO_x emission factor by matching TROPOMI derived NO_x emissions with concurrent MODIS FRP, which is considered as a good indicator of fire intensity (Ichoku and Kaufman, 2005; Wiggins et al., 2020). Since the lifetime of NO_x is within several hours (Figure 5), the influence of fire emissions from previous days should be negligible. We have clarified the limitation of TROPOMI as follows:

TROPOMI is limited to single overpass per day, which cannot resolve the short-term evolution of fire plumes. The newly launched or upcoming geostationary satellite instruments such as GEMS and TEMPO will offer an unprecedented opportunity to continuously observe the emissions and chemical evolution of NO_x from fires that will no longer be limited to a single snapshot (Chance et al., 2013; Kim et al., 2019).

Specific comments.

1. Line 11: "behaviour" should be "behavior". "occur" should be "that occur".

Reply: Done.

2. Line 15: The sentence is a little confusing. I think the authors recalculate the NO2 VCD of every pixel with the GEOS-CF simulated profile not only over the fire plumes?

Reply: It's true that we apply the GEOS-CF profile for every pixel, but overall such replacement only increases the NO_2 VCD for plume affected pixels. We have revised the sentence as follows:

We update the *a priori* profile of NO_2 with a fine-resolution (0.25˚) global model simulation from NASA's GEOS Composition Forecasting System (GEOS-CF), which largely enhances NO_2 columns over fire plumes.

3. Line 24 and 27: Please list enough examples when you give examples.

Reply: We have added more examples as suggested:

Biomass burning emissions affect global radiative forcing, the hydrological cycle, ecosystem and air quality (e.g., Crutzen and Andreae, 1990; Penner et al., 1992; Johnston et al., 2012; Liu et al., 2014).

Biomass burning emissions inventories used in models are subject to uncertainties in estimates or measurements of the burned area, fuel loadings, combustion efficiency, and also the compound-specific emission factors that relate the mass of a chemical species emitted to fuel consumption (e.g., Petrenko et al., 2012; Liu et al., 2020; Carter et al., 2020).

4. Line 42: "the fire detection".

Reply: Done.

5. Line 44: "has a finer spatial resolution".

Reply: Done.

6. Line 46: "spatial resolutions"

Reply: Done.

7. Line 57 and 64: Please cite recent and more relevant studies about TROPOMI.

Reply: We have added more recent studies about TROPOMI:

The accuracy of satellite retrieval of NO_2 columns largely depends on the *a priori* knowledge of NO_2 vertical profile shape needed for calculating air mass factor (e.g., Boersma et al., 2018; Verhoelst et al., 2021).

Replacing the *a priori* vertical profile from a fine-resolution regional model can enhance the spatial gradient and correct the low bias of satellite retrieved NO_2 over polluted regions (e.g., Russell et al., 2011; Valin et al., 2011; Goldberg et al., 2017; Ialongo et al., 2020; Judd et al., 2020; Tack et al., 2021).

8. Line 83: "afternoon global" is quite obscure, please specify the overpassing time is around 13:30 local time.

Reply: Done. Revised as follows:

TROPOMI provides afternoon (~ 1:30 PM local time) global observations in the UV−visible−near infrared−shortwave spectra with a fine spatial resolution of 7×3.5 km^2 at nadir (increased to 5.5×3.5 km^2 since August 2019).

9. Line 86-93: Do you use the S5P operational product that retrieved by KNMI? If so, you should cite van Geffen et al., (2019) when introducing the way of retrieval. Besides, you should also cite the validation paper (i.e. Tijl et al., 2021 https://amt.copernicus.org/articles/14/481/2021/) when discussing the underestimation of S5P.

van Geffen, J. H. G. M., Eskes, H. J., Boersma, K. F., Maasakkers, J. D., and Veefkind, J. P.: TROPOMI ATBD of the total and tropospheric NO2 data products (issue 1.4.0), Royal Netherlands Meteorological Institute (KNMI), De Bilt, the Netherlands, 2019.

Reply: We have added the suggested references.

10. Line 165: The format of the reference is "Laughner and Cohen, (2019)".

Reply: Done.

11. Line 203: Not very clear to me why "3 to 30 days before and after the fire day" is 56 days in total.

Reply: We include the days 30 days before and after the fire day, which is 60 days, and we further exclude two days before and after the fire because fire often lasts for several days, which gives 56 (i.e., 60 – 4 = 56) days in total. We have revised the sentence as follows:

We then select fires where TROPOMI NO_2 tropospheric columns on the fire day are at least one standard deviation higher than the mean TROPOMI NO_2 columns 30 days before and after the fire day (excluding the nearest four days as fires may last for several days, defined as Ω_{NO2_B}).

12. Line 206: "filter" should be "filters".

Reply: Done.

13. Line 224-225: Please give specific examples to explain "We also exclude fires in which TROPOMI NO2 line densities are monotonically increasing or decreasing within the region."

Reply: We have revised the sentence as follows:

We only include fires in which TROPOMI NO_2 line densities peak near the fire centre, meaning that fires with monotonically increasing or decreasing line densities within the region are excluded.

14. Line 241: "10000 MW" should be "10,000 MW"

Reply: Done.

15. The resolution of Figure 2 is too low. It's better to start with the original TROPOMI NO2 data before (a).

Reply: We provide high-resolution figures for the revised manuscript. The resolution or the re-gridded data is 0.05˚ for the maps shown in Figure 2, which is close to the original resolution of TROPOMI (5.5 × 3.5 km^2). We did not start with the original TROPOMI NO2 data because the first step of processing TROPOMI data is to rotate swaths data along the wind direction. The original TROPOMI data can be found from TEMIS website (https://www.temis.nl/airpollution/no2col/). We have added a supplementary figure that compares TROPOMI and OMI data for the fire episode:

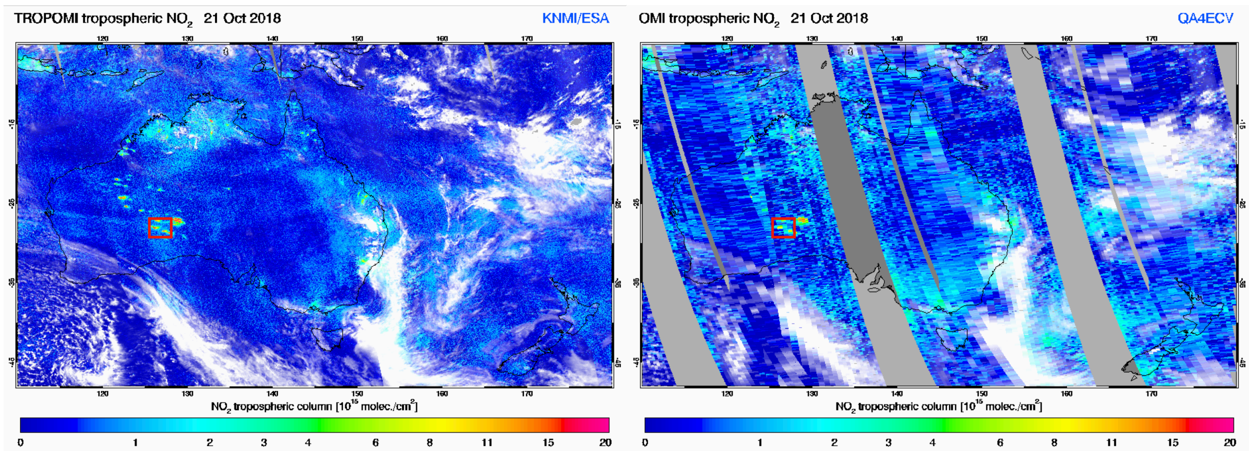

Figure S3 Maps of TROPOMI (left) and OMI (right) tropospheric NO_2 over Australia on October 21, 2018. The figures are acquired from TEMIS: https://www.temis.nl/airpollution/no2.php. The red box labels the location of the fire episode shown in Figure 2.

References:

Ichoku, C. and Kaufman, Y. J.: A Method to Derive Smoke Emission Rates From MODIS Fire Radiative Energy Measurements, Ieee T Geosci Remote, 43, 2636–2649, https://doi.org/10.1109/tgrs.2005.857328, 2005.

Wiggins, E. B., Soja, A. J., Gargulinski, E., Halliday, H. S., Pierce, R. B., Schmidt, C. C., Nowak, J. B., DiGangi, J. P., Diskin, G. S., Katich, J. M., Perring, A. E., Schwarz, J. P., Anderson, B. E., Chen, G., Crosbie, E. C., Jordan, C., Robinson, C. E., Sanchez, K. J., Shingler, T. J., Shook, M., Thornhill, K. L., Winstead, E. L., Ziemba, L. D., and Moore, R. H.: High Temporal Resolution Satellite Observations of Fire Radiative Power Reveal Link Between Fire Behavior and Aerosol and Gas Emissions, Geophys Res Lett, 47, https://doi.org/10.1029/2020gl090707, 2020.