Reply to Reviewer 2

This paper estimates biomass burning NOx emissions and lifetime using daily observations from TROPOMI. The topic has a broad interest, and the investigation is solid. I particularly appreciate the validation using a plume model. The work suggests decreasing NOx lifetime with fire intensity due to the increase in both NOx abundance and hydroxyl radical production. I would recommend minor revision before publication.

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.

General comments.

1. Section 3.1 profile correction using GEOS-CF: How sensitivity the derived lifetime and emissions to this correction?

Reply: We have shown in Sect. 4.3 that using GEOS-CF profiles largely enhances the NO₂ column density and the derived emissions. Figure 4 shows that using updated profile increases the derived emission factors. Figure S5 shows the relationship between emissions and FRP using original TROPOMI NO₂ data without profile correction. The derived NO_x lifetime is not sensitive to the profile correction, largely because the lifetime is determined by the shape of the fire plumes that are not affected by the *a priori*. We have added a new supplementary figure that shows the derived NO_x lifetime using the original TROPOMI data:



Figure S7 Same as Figure 5 but using original TROPOMI NO₂ data without updating the a priori profile.

We have added the following discussions on the sensitivity of derived NO_x lifetime to profile correction:

We find similar NO_x lifetime using original TROPOMI NO₂ data, largely because the derived NO_x lifetime is determined by the shape of fire plumes that are not affected by the *a priori*.

2. Section 3.2. It is not very clear to me how ALH is related to the EMG approach. I suppose the authors indicate a consistent wind layer height and injection height. If so, I would suggest making this clearer in the text. Are the derived results from the EMG approach very sensitive to the choice of wind layer heights? Additional sensitivity analysis would be beneficial to the study.?

Reply: We'd like to clarify that we did **not** use a consistent wind layer height. ALH is used to determine the fire injection height. Assuming consistent wind layer height generally works fine for small fires, but big fires are often associated with high injection height. We clarify this in the main text:

Previous studies either use the averaged wind speed of the first several layers (Beirle et al., 2011; Lu et al., 2015) or choose a constant layer such as 900 hPa (Mebust et al., 2011), but injection height of wildfires varies significantly, especially for large fires which inject emissions into high altitudes (Val Martin et al., 2010). To account for varying injection height, we use TROPOMI ALH as an approximation of the fire injection height instead of assuming a constant layer. We vertically interpolate ERA-5 wind data to the pressure level of aerosol layer. For the fires without valid ALH (~36% of the selected fires), we use 900hPa, as the ALP level for the majority of selected fires is near 900 hPa (see Sect. 4.1).

We have conducted sensitivity analysis in Section 5.3. We estimate that an increase of 500 m ALH corresponds to ~22% increase of the wind speed on average, meaning that NO_x lifetime will decrease by ~18%. In Sect. 5.3, we have discussed how EMG approach is sensitive to the choice of wind layer heights:

Uncertainty and variance of the wind speed, however, should lead to errors in the derived NO_x lifetime. Here we determine the wind speed by interpolating the wind profile to TROPOMI derived aerosol layer. Comparison of TROPOMI ALH with plume height from the Cloud-Aerosol LIdar with Orthogonal Polarization (CALIOP) and the Multi-angle Imaging SpectroRadiometer (MISR) measurements suggest that TROPOMI ALH is overall 500 m lower (Griffin et al., 2020; Nanda et al., 2020). We estimate that an increase of 500 m ALH corresponds to ~22% increase of the wind speed on average, meaning that NO_x lifetime will decrease by ~18%.

3. Section 3.4. "pixels are grouped to separate plumes based on their connections with surrounding pixels" I recommend a diagram or plot here to illustrate the grouping algorithm. It is not easy for me to get it from the text here.

Reply: We have added a figure in Supplement to illustrate the processes of the grouping algorithm:



Figure S2 Illustration of the processes that identify and filter out nearby plumes.

4. Section 4.5. I'm surprised to see the results for the wind speed less than 2 m/s. As far as my understanding, the EMG function is not suitable for the cases of calm winds. Is there any special reason for applying EMG for calm-wind conditions?

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 (< 4%). 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.

Specific comments.

5. Page 2, line 49. Please add reference for "the improved signal-to-noise ratio". I would

suggest more details about the improved signal-to-noise ratio to justify the usage of daily observation. For example, how does the ratio improve from OMI to TROPOMI? How does one TROPOMI observation compare to several OMI observations? It is not necessary to discuss this in the abstract, but somewhere in the main text would be appreciated.

Reply: We have added references for TROPOMI:

The finer spatial resolution (\sim 7 × 3.5 km²), and the improved signal-to-noise ratio of TROPOMI compared to OMI offer new opportunities to more reliably interpret observations of individual plumes (Veefkind et al., 2012; Judd et al., 2019; van Geffen et al., 2020).

We also include a new supplementary figure that directly compares daily TROPOMI vs. OMI observation for detecting fire NO_x . The figure clearly shows the improved performance of TROPOMI over OMI:



Figure S4 Maps of TROPOMI (left) and OMI (right) tropospheric NO₂ over Australia on October 21, 2018. The figures are acquired from TEMIS: https://www.temis.nl/airpollution/no2col/. The red box labels the location of the fire episode shown in Figure 2.

We have added the following discussions in the main text:

Several NO₂ plumes are detected by TROPOMI on this day, which outperforms OMI observation on the same day which detects less smaller fires, shows less spatial gradients and larger data gap (Figure S4).

6. Page 6, line 180. Please add reference for PECANS. Additionally, please clarify the reasons for the settings in the model, such as the diffusion coefficients and O3 concentrations.

Reply: We have added reference for PECANS:

To understand the factors that control the NO_x lifetime, we employ a one-dimensional (1-D) multi-box plume model based on the Python Editable Chemical Atmospheric Numerical Solver (PECANS; Laughner and Cohen, 2019; Laughner 2019).

We have clarified the reasons for settings:

The wind speed is fixed at 5 m/s, and the diffusion coefficients are also fixed at 100 m^2/s following Laughner and Cohen (2019).

The O₃ concentration is fixed at 65 ppbv, which is close to observed mean O₃ concentration near fire plumes (Alvarado and Prinn, 2009; Alvarado et al., 2014). A fixed branching ratio to form RONO₂ in RO₂ + NO reaction of 0.05 is used following Laughner and Cohen (2019).

7. Line 227. Please clarify the details of the 50 initial conditions.

Reply: We have revised the sentence as follows:

To test the sensitivity of the fitting results to initial conditions, we repeat the fitting with varying initial values for each parameter 50 times, and we exclude fires where the standard deviation of resulting emissions is more than 50% of the emissions.

8. Line 305. Does Mebust and Cohen (2014) adopt a similar method as this study? If not, I would suggest rephrasing this part by mentioning the results using standard TROPOMI products firstly and then comparing with that of Mebust and Cohen (2014). Otherwise, the readers may get confused here.

Reply: Mebust and Cohen (2014) use different method as this study. We have revised this part to avoid confusion:

Using the standard TROPOMI NO₂ 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 NO₂ with *in situ* measurements also suggest TROPOMI NO₂ 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 NO₂ in Mebust and Cohen (2014). Using the standard TROPOMI NO₂ 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.

8. Section 5.3. please clarify the calculation of chemical lifetime.

Reply: We have revised the sentence as follows:

To assess if EMG fitted NO_x lifetime is indicative of the chemical lifetime, we use the PECANS model to calculate an EMG fitted lifetime and a chemical lifetime of NO_x from

two permanent losses of NO_x through the formation of HNO_3 and $RONO_2$ over downwind region (*i.e.*, mean NO_x concentration divided by the mean chemical loss of NO_x).