

## Point-by-point Responses to Reviewer #2 for # MS No.: acp-2022-447

The authors have used regular fonts for the Referee's comments (which might be divided into two or multiple comments), blue fonts for our responses, and red fonts with quotation marks to show the revised text.

### Reviewer 2 # Comments

Overall, this is an excellent research article. In addition to the suggested revisions of the other reviewer, I would like to see improvement in the abstract as well as the summary and conclusions, which are a bit thin on the important implications of your research. For instance, you make the following statement in the summary and conclusions: "Our study on both savanna and temperate forest fire emissions demonstrates the capability and limitations of TROPOMI data for the study of the regional variability of combustion characteristics and their impacts on regional atmospheric composition and air quality." You make a similar comment in the abstract. This statement may be accurate, but I would like you to elaborate on this statement, including on how your technique may be applied to other world regions. As another example, you say: "These differences could be traced back to different measurement techniques used, their spatial resolutions, nonlinear sensitivities to gas densities in the boundary layer, and larger NO<sub>2</sub> natural variability due to its short lifetime, all of which suggest that further validation of satellite products and investigations of more cases are required." Could you suggest additional validation that would be most helpful to this end? How many cases are required? My recommendation is to revisit the abstract and summary and conclusions with an eye for elaborating on the broader implications of your research.

**Response:** Thank you for your comments. We rewrote the abstract,

"The bushfires that occurred in Australia in late 2019 and early 2020 were unprecedented in terms of their scale, intensity, and impacts. Using nitrogen dioxide (NO<sub>2</sub>) and carbon monoxide (CO) data measured by the Tropospheric Monitoring Instrument (TROPOMI), together with fire counts and fire radiative power (FRP) from MODIS, we analyzed the temporal and spatial variation of NO<sub>2</sub> and CO column densities over three selected areas covering savanna and temperate forest vegetation. The  $\Delta\text{NO}_2/\Delta\text{CO}$  emission ratio and emission factor were also estimated. The  $\Delta\text{NO}_2/\Delta\text{CO}$  emission ratio was found to be  $1.57 \pm 1.71$  for temperate forest fire and ranged from  $2.0 \pm 2.36$  to  $2.6 \pm 1.92$  for savanna fire. For savanna and temperate forest fires, satellited-derived NO<sub>x</sub> emission factors are  $1.48 \text{ g kg}^{-1}$  and  $2.39 \text{ g kg}^{-1}$  separately, while their CO emission factors are from 107.39 to 126.32  $\text{g kg}^{-1}$ . This study demonstrates that the large-scale emission ratios derived from the TROPOMI satellite for different biomass burnings can help identify the relative contribution of smoldering and flaming activities in a large region and their impacts on the regional atmospheric composition and air quality. This method can be applied to study the emissions from other large fires, or even the burning of fossil fuel in megacities, and their impact on air quality."

Accordingly, we also rewrote the summary and conclusion in our revision as,

"The 2019-2020 black summer fires in Australia emitted large amounts of trace gases and aerosols. In this study, we focused on the analysis of two trace gases: CO and NO<sub>2</sub>. Based on the total columns (mean and maximum) from TROPOMI observations and the fire intensity from MODIS in August 2019 to early 2020, we estimated the ERs of NO<sub>2</sub> relative to CO for each day over three selected areas with savanna and temperate forest vegetation. For temperate forest fires, the ER was  $1.57 \pm 1.71$  which is consistent with previous studies. For savanna vegetation fires, the ER ranged from  $2.0 \pm 2.36$  to  $2.6 \pm 1.92$ , which was slightly lower compared to other studies. These differences could be traced back to different

measurement techniques used, their spatial resolutions, nonlinear sensitivities to gas densities in the boundary layer, and larger NO<sub>2</sub> natural variability due to its short lifetime, all of which suggest that further validation of satellite products and investigations of more cases are required. For example, aircraft measurements from NASA airborne campaigns could be used to validate TROPOMI satellite-derived CO and NO<sub>2</sub> concentrations. The satellite-derived concentrations and emission can be also compared with simulations from dynamical models (e.g., Weather Research and Forecasting model coupled to Chemistry, Community Modeling and Analysis System). Further advanced techniques to improve the calibration and retrieval algorithm could be used to improve the estimates of emissions and emission factors. For instance, even though we used the improved TROPOMI NO<sub>2</sub> data from van Geffen et al. (2022) in this study, it still has a negative bias when compared with ground-based observations, which probably is due to the relatively coarse resolution (1°×1°) of the *a priori* profiles used. Taking advantage of higher-resolution profile shapes can lead to better retrieval of tropospheric columns over emission hotspots (Douros et al., 2022). Additionally, considering the short lifetime of NO<sub>2</sub>, the NO<sub>2</sub> tropospheric column could be corrected using boundary layer temperature and OH concentration, as described in the work of Lama et al. (2019).

Using the methods from Mebust et al. (2011) and Adams et al. (2019), net emission fluxes were estimated by using a 14-day CO effective lifetime and a 2.5-hour NO<sub>2</sub> effective lifetime, and EFs were calculated. The TROPOMI-derived NO<sub>x</sub> EFs were 1.48 g kg<sup>-1</sup> and 1.51 g kg<sup>-1</sup> for savanna and temperate forest fires which are lower than previous studies while the CO EFs were 107.39 g kg<sup>-1</sup> for savanna fires and 136.41 g kg<sup>-1</sup> for the temperate forest. Our study on both savanna and temperate forest fire emissions demonstrates the capability and limitations of TROPOMI data for the study of the regional variability of combustion characteristics and their impacts on regional atmospheric composition and air quality. Benefiting from the global coverage of TROPOMI and its high spatial resolution, the method used in our study could be applied to different vegetation wildfires at various scales, even the burning of fossil fuel in megacities.”

## Reference

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