

Dear Editor Dr. James Lee,

Thank you very much for handling our manuscript. Please find below our itemized responses to the reviewers' comments and a marked-up manuscript. We have addressed all the comments raised by the reviewer and incorporated them in the revised manuscript.

Thank you for your consideration.

Sincerely,
Haolin Wang et al.

Reviewer #1

Comment [1-1]: Wang et al. aim to enhance the temporal resolution of current biomass burning emission estimates, which are typically provided on a daily to monthly basis. When integrated into chemical transport models, these emission estimates currently rely on fixed diurnal cycles informed by climatological empirical data. The authors introduce a new method that utilizes day-specific diurnal variations derived from geostationary Fire Radiative Power (FRP). They then examine how these improvements in emissions impact simulated ozone levels. Overall, the subject matter aligns with the scope of ACP. The presentation is well-structured and clear, though the resulting improvements are subtle.

Response [1-1]: We thank the reviewer for the positive comments on our study. Below, we provide a point-by-point response to the reviewer's comments and summarize the changes that have been made in the revised manuscript.

Comment [1-2]: Abstract, L21-23: The authors' new method of incorporating day-specific diurnal variations from geostationary observations, compared to fixed diurnal cycles based on climatological data, appears to yield only subtle improvements. The surface ozone biases are reduced slightly from '−1.54 to +9.09 ppbv' to '−1.58 to +9.13 ppbv'. This is particularly noteworthy given that significant changes to ozone levels can be achieved by addressing other uncertainties in wildfire emissions (e.g., injection height, emission budgets, etc.). The diurnal cycles of NO_x seem to be reasonably captured by the diurnal scaling factors applied in both the GFED4 and GFAS inventories (Fig. 1), with no significant differences in bias between the new method and the conventional GFED and GFAS approaches (Fig. 3). How does this new method improve model performance or enhance our general understanding of current knowledge?

Response [1-2]: We thank the reviewer for this constructive comment. We agree that the overall statistical improvement in surface ozone biases is small. However, the principal contribution of our approach is not simply to reduce the bias, but to provide a more physically realistic representation of the diurnal variability of biomass burning emissions. Our simulations show that the timing and amplitude of diurnal NO_x emissions differ substantially between the geostationary-derived inventory and conventional climatological scaling factors. While these differences do not strongly alter domain-wide mean biases due to the smoothing effect of long-range transport, and the fact that most available evaluation datasets are located outside the core African fire regions, they lead to pronounced local and regional impacts on ozone (up to −8.57 to +21.88 ppbv at the surface and −0.41 to +1.09 DU in tropospheric columns) and on OH concentrations (−4.4% to +51.7%) during the fire season. These effects propagate to remote regions through atmospheric circulation, influencing chemical regimes as far

afield as South America and the Persian Gulf. Thus, our method enhances our understanding of how sub-daily fire activity modulates atmospheric chemistry and demonstrates the importance of integrating realistic diurnal variability in future global models. We have revised the text to highlight these mechanistic insights in addition to the statistical evaluation as follows.

L22–26: “Simulations with real hourly-resolved emissions produce comparable overall surface ozone biases (–1.54 to +9.09 ppbv vs. –1.58 to +9.13 ppbv) and marginally higher correlations with TROPOMI nitrogen dioxide ($r = 0.80$ – 0.89) and OMI ozone ($r = 0.80$ – 0.94). Although the statistical improvements are limited, the geostationary-driven approach reveals pronounced regional ozone differences and mechanistic insights into the role of diurnal fire variability.”

L477–481: “This insensitivity partly reflects the fact that most available evaluation datasets are located outside the core fire regions of Africa, where long-range transport tends to smooth local diurnal variations. As a result, bulk evaluation metrics underestimate the impact of sub-daily emission differences. Nevertheless, our sensitivity experiments demonstrate that diurnal variability exerts important mechanistic influences on ozone chemistry and transport that are not captured by fixed empirical diurnal cycles.”

L684–686: “This limited improvement likely stems from the smoothing effect of long-range atmospheric transport, which diminishes the impact of high-resolution diurnal variations, and the fact that most available evaluation datasets are located outside the core African fire regions, where local diurnal signatures are strongest.”

Comment [1-3]: L229: Unclear how CO relates to other emitted species. By a fixed CO-to-species ratio for all three emission inventories? This is an important point to clarify before interpreting the results in Section 3.1 where intra-inventory NO_x emissions are compared.

Response [1-3]: We thank the reviewer for this valuable comment and agree that the distinction between the inventories requires clearer explanation. In our study, the treatment of CO and other species differs among the three biomass burning inventories as follows:

- **FREM:** The FREM inventory provides top-down estimates of CO emissions based on geostationary SEVIRI FRP and Sentinel-5P CO observations (Nguyen et al., 2023). Other species (e.g., NO_x, VOCs, aerosols) are derived from CO using biome-specific CO-to-species emission factor ratios reported in Andreae and Merlet (2001), Akagi et al. (2011), and Andreae (2019), consistent with Nguyen et al. (2023). This approach ensures that the conversion accounts for differences among vegetation types (e.g., savanna, tropical forest).
- **GFED4.1s:** GFED is a bottom-up inventory that combines MODIS burned area, fuel load estimates, combustion completeness, and biome-specific emission factors defined per unit of dry matter burned (Randerson et al., 2017; van der Werf et al., 2017). Therefore, it directly provides emissions for multiple species, which we use without any rescaling from CO.
- **GFAS:** GFAS assimilates MODIS FRP observations and converts them to dry matter combustion using land-cover-specific FRP-to-dry-matter-consumed factors that relate MODIS FRP to dry matter combustion rates as in GFED (Kaiser et al., 2012). It then estimates emissions of trace gases and aerosols using land-class-dependent emission factors.

Accordingly, we have revised Section 2.5 (Lines 278–285) to explicitly clarify this distinction as follows: “In GFED4.1s, emissions of trace gases and aerosols are derived using a bottom-up approach that combines MODIS

burned area, fuel load, combustion completeness, and biome-specific emission factors defined per unit of dry matter burned (Randerson et al., 2017; van der Werf et al., 2017). GFAS follows a similar principle, converting MODIS FRP into dry matter combustion using land-cover-specific FRP-to-dry-matter-consumed factors, and then deriving emissions using biome-dependent emission factors (Kaiser et al., 2012). By contrast, the FREM inventory provides only CO emissions derived from geostationary FRP and Sentinel-5P CO observations (Nguyen et al., 2023); other species (e.g., NO_x, VOCs, aerosols) are obtained by scaling CO using biome-specific CO-to-species emission factor ratios (Andreae and Merlet, 2001; Akagi et al., 2011; Andreae, 2019)."

References

- Andreae, M. O., and Merlet, P.: Emission of trace gases and aerosols from biomass burning, *Global Biogeochem. Cycles*, 15, 955–966, 2001.
- Akagi, S. K., et al.: Emission factors for open and domestic biomass burning for use in atmospheric models, *Atmos. Chem. Phys.*, 11, 4039–4072, 2011.
- Andreae, M. O.: Emission of trace gases and aerosols from biomass burning – an updated assessment, *Atmos. Chem. Phys.*, 19, 8523–8546, 2019.
- Kaiser, J. W., et al.: Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power, *Biogeosciences*, 9, 527–554, 2012.
- Randerson, J. T., et al.: Global Fire Emissions Database, Version 4.1 (GFEDv4), ORNL DAAC, 2017.
- van der Werf, G. R., et al.: Global fire emissions estimates during 1997–2016, *Earth Syst. Sci. Data*, 9, 697–720, 2017.
- Nguyen, H. M., He, J., and Wooster, M. J.: Biomass burning CO, PM and fuel consumption per unit burned area estimates derived across Africa using geostationary SEVIRI fire radiative power and Sentinel-5P CO data, *Atmos. Chem. Phys.*, 23, 2089–2118, 2023.

Comment [1-4]: L300 & L567: The authors may want to clarify the intra-inventory differences in the species budgets for CO, PM, and other species, in addition to the NO_x presented. This would help readers better understand the full context of the intra-inventory differences and make sense of the comparisons being made.

Response [1-4]: We thank the reviewer for this helpful suggestion. In the main text we focus on NO_x because of its direct role in ozone formation, while CO is the principal species used for intercomparison across multiple biomass burning emission inventories and serves as a key tracer of combustion and long-range transport. To provide a broader context, we have therefore included an analysis of the intra-inventory differences for CO in the Supplement (Figure S1). The CO results exhibit similar patterns to those of NO_x, supporting the robustness of our conclusions.

We have clarified in the text: “A similar comparison for CO emissions is presented in Figure S1. CO is the principal species used for intercomparison across multiple biomass burning emission inventories (Pan et al., 2020; Jin et al., 2024) and serves as a key tracer of combustion and long-range transport (Duncan et al., 2003; Edwards et al., 2006). All three inventories capture the pronounced north–south seasonal contrast of African fires, with peak activity in January and July and relatively weak emissions in April and October. The total emitted CO differs noticeably among inventories, with GFED4 generally reporting higher regional totals than the others, while the relative differences vary somewhat between months and regions. The diurnal variation of CO emissions is broadly consistent with that of NO_x, showing a clear daytime maximum around local noon and minima at

night, but the amplitude and peak timing vary among inventories, reflecting differences in the representation of fire activity and emission factors.”

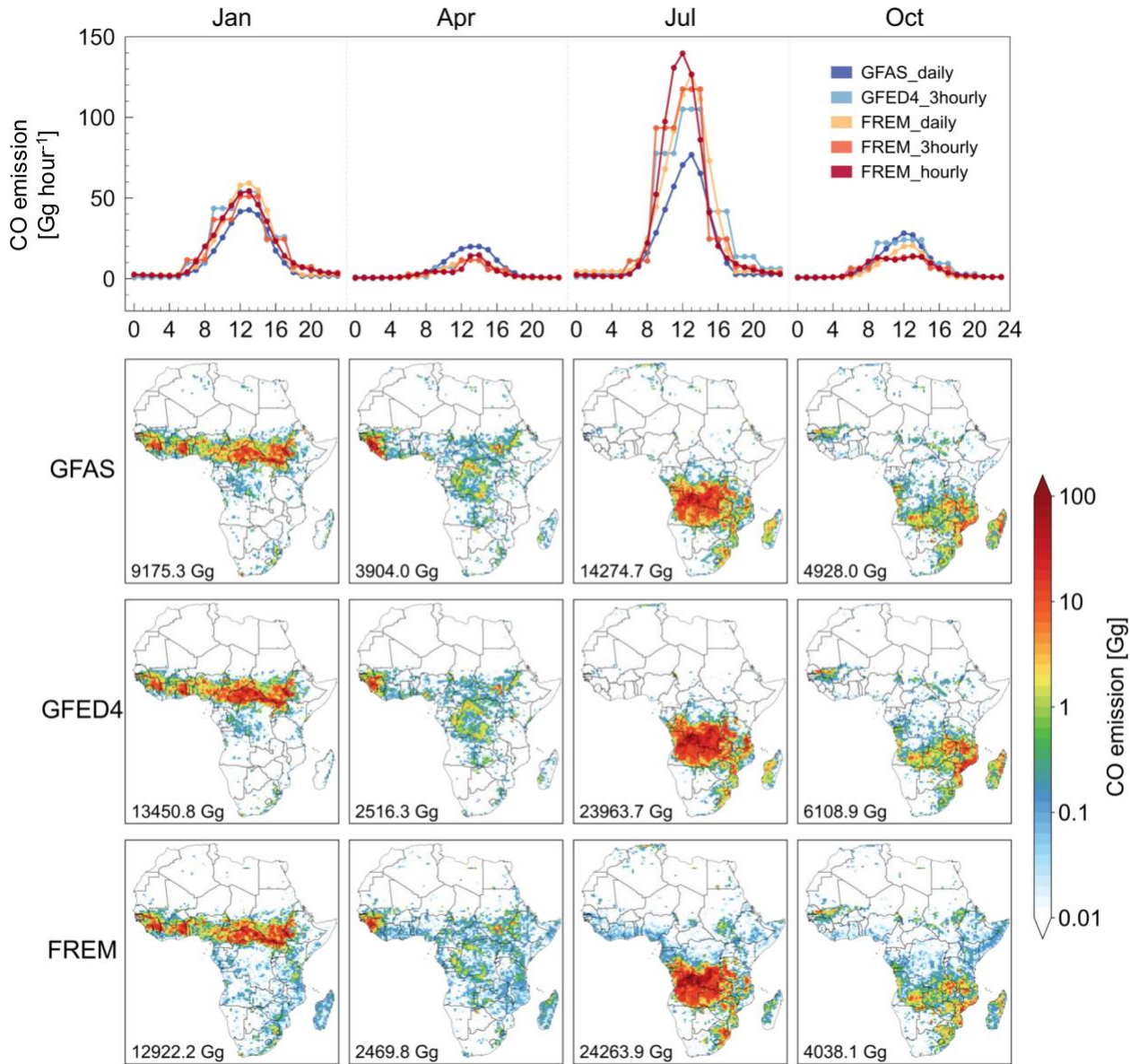


Figure S1. Same with Figure 1, but for CO emission.

Comment [1-5]: Section 3.4: Most global effects are minor, around ± 0.1 -1 ppbv in Fig.8.

Response [1-5]: We thank the reviewer for this comment. We agree that most global mean surface ozone changes shown in Fig. 8 are relatively small (typically ± 0.1 –1 ppbv). This reflects the fact that the diurnal variability of African fire emissions is a sub-daily redistribution of the same daily emission totals, so the largest impacts are expected locally and seasonally rather than in global mean fields. Nevertheless, our simulations show that in regions with intense fire activity (e.g., Angola and Zambia) and along major outflow pathways, the differences are much larger (up to 7–8 ppbv at the surface and ± 0.4 DU in tropospheric columns). Furthermore, even small global-scale changes of ~ 0.5 ppbv can be significant when integrated over the free troposphere, where

they affect the oxidative capacity and radiative forcing of ozone. We have revised the text in Section 3.4 to clarify that while global mean effects are generally modest, the regional and process-level impacts are substantial and provide the main scientific value of including realistic diurnal variability.

We have clarified in the text: “These variations also influence downwind regions along major transport pathways, altering ozone precursors and oxidative capacity far beyond Africa, potentially impacting trend interpretations (Wang et al., 2024). Importantly, even small changes at the global scale can be relevant for the tropospheric ozone budget and radiative forcing. Thus, the main scientific value of including geostationary-derived diurnal variability lies in capturing these regional and process-level impacts rather than producing large global mean differences.”

Comment [1-6]: Conclusion: The new method may be more significant for understanding the hourly variations in atmospheric chemistry that occur throughout the diurnal progression of large wildfires. However, for general wildfire simulations, the improvements in the model are less noticeable.

Response [1-6]: We thank the reviewer for this helpful suggestion. We agree that the overall improvements in bulk model performance for general wildfire simulations are modest. However, the primary value of our method lies in providing a more physically realistic description of the diurnal variability of fire emissions, which is particularly important for understanding the hourly progression of atmospheric chemistry during large wildfire events.

We have clarified in the text: “Moreover, our findings suggest that simulations of BB emissions should consider their diurnal variations to assess their impact more accurately on atmospheric chemistry. While the improvements in bulk model performance for general wildfire simulations are modest, the main significance of our method lies in capturing hourly variability and advancing process-level understanding of atmospheric chemistry during large wildfire events.”

Comment [1-7]: Title: Define the acronym FRP.

Response [1-7]: We thank the reviewer for this comment. We have revised the title.

Comment [1-8]: Fig.4: Define the acronym REF in the figure caption.

Response [1-8]: We thank the reviewer for pointing this out. We have revised the caption of Fig. 4 to define the acronym.

We have clarified in the text: “The REF point on the horizontal axis denotes the reference observations, representing perfect agreement with normalized standard deviation = 1, correlation coefficient = 1, and centred RMSE = 0.”