

**Editor:**

*Thank you for your revisions and detailed responses to the first round of reviews. Both reviewers acknowledge that your substantial efforts have addressed many of their technical questions.*

*I have now received the second-round reviews, and while both reviewers recognize your improvements, they also identify remaining issues that they believe should be further addressed. I share the view that these concerns are substantial enough to require further revision.*

*From my perspective, the reviewers' concerns are grouped into three areas:*

1. Observational uncertainty and retrieval biases.

*Have the uncertainties in your inversion framework been fully characterized, particularly regarding the satellite XCO retrievals? They both are concerned whether retrieval uncertainties may be larger than assumed, not random, or even biased due to co-emitted aerosols under high-AOD conditions.*

2. Representativeness of sampled fires.

*While your sensitivity tests demonstrate robustness of the season-total emissions, questions remain about whether the inversion framework can adequately constrain low-intensity or diffuse fire emissions. Furthermore, whether the system can identify missing sources (including large fires) when they are absent from the prior. Not being able to scale a zero prior is a known limitation in this type of inversion.*

3. Broader literature context.

*The paper may benefit from a wider engagement with recent work on observationally constrained estimates of emissions, atmospheric composition, and radiative effects.*

*For these reasons, I believe that a careful revision would substantially strengthen the paper.*

*I look forward to seeing your revised manuscript.*

We thank the editor for the careful assessment of our revised manuscript and for providing a clear summary of the concerns that were raised. We are grateful for the constructive feedback from both reviewers and appreciate their kind recognition of revision efforts thus far. We have carefully considered and addressed the latest feedback.

**Reviewer II:**

*The authors have made substantial revisions in response to the first-round comments. The additions of the 100/150 ppb threshold sensitivity tests (Sect. 2.5.4), the free-tropospheric bias experiment (Appendix A3), the quantification of MOPITT data gaps (Sect. 3.3), and the 6-day rolling correlation analysis (Appendix A6) are valuable contributions that address my RC2 concerns. I am generally satisfied with the responses to my original 11 comments, particularly the new quantitative analyses for comments 1, 3, 5, 8, and 9. The remainder of this review focuses on a few issues that I noticed upon reading the revised manuscript, which I believe warrant further attention before the manuscript is finalized.*

We would like to thank the reviewer for the constructive feedback and carefully evaluating our revised manuscript, and are glad to hear the previous responses were satisfactory. We appreciate the reviewer's engagement by raising the points related to aerosols effects and small-fire constraints. Please find our more detailed responses to the individual comments are below, together with descriptions of manuscript changes that were made in response to the reviewer's suggestions.

*The most important issue I would like to raise is the absence of any discussion of aerosol interference on the satellite XCO retrievals. The 125 ppb XCO threshold used for fire plume selection necessarily identifies the most intense burning conditions, which are also the conditions under which aerosol optical depths are highest. Both TROPOMI SWIR and MOPITT TIR retrievals are known to be susceptible to aerosol-induced biases under high-AOD conditions, and the direction of these biases depends on aerosol type, vertical distribution, and single-scattering albedo. The manuscript applies a quality filter (TROPOMI qa\_value > 0.5) and inflates the model-data mismatch (Sect. 2.5.4), but neither step specifically addresses retrieval bias caused by aerosols in dense fire plumes. The free-tropospheric bias sensitivity experiment in Appendix A3 tests the impact of a uniform 20 ppb CO bias and is informative for model-related errors, but it does not speak to whether the satellite retrievals themselves are biased under the very conditions selected for assimilation. I would appreciate seeing at least a discussion of the potential direction and magnitude of aerosol-induced biases in the TROPOMI and MOPITT XCO retrievals under high-AOD fire plume conditions. An examination of the AOD distribution associated with the assimilated ( $XCO \geq 125$  ppb) observations, cross-referenced with independent aerosol products such as MODIS AOD, would be ideal, but even a qualitative assessment of whether aerosol effects would lead to systematic over- or underestimation of XCO, and thus of inferred fire emissions, would materially strengthen the manuscript.*

We thank the reviewer for raising this important point. Aerosol-induced retrieval errors are indeed a potential concern when

assimilating CO observations in wildfire plumes because aerosols modify the light path of photons which might affect retrieval sensitivity. The reviewer is correct in pointing out that in our effort to isolate observations associated with fire signals, some observations we feed to the inversion system might be associated with enhanced aerosol loads.

For TROPOMI, multiple scattering effects are explicitly considered within the SWIR retrieval through the retrieval of effective light-path/scattering parameters, supported by simultaneous fitting of CH<sub>4</sub> and H<sub>2</sub>O absorption features. Sensitivity studies presented in the TROPOMI algorithm theoretical baseline document indicate that residual biases remain below approximately 1% across a range of aerosol scenarios, including polluted conditions (Landgraf et al., 2024). Furthermore, Rowe et al. (2022) evaluated TROPOMI CO retrievals within optically thick wildfire smoke plumes during a targeted field campaign and found that retrieval performance remained within the mission accuracy (15% accuracy, 10% precision) requirements.

Please note that, to some degree, we filter out retrievals under potentially problematic observing conditions through applying the (qa\_value > 0.5). More specifically, this means we do not include retrievals when the retrieved aerosol optical thickness and/or cloud heights are  $\geq 0.5$ , and  $\geq 5\text{km}$  respectively, or  $\leq 0.5$  and  $\geq 500\text{m}$  respectively (Landgraf et al., 2025). However, we acknowledge that this does not cover all smoke plumes.

Aerosol radiative effects in the thermal infrared (TIR) are weak (e.g. Eck et al., 1999), while the MOPITT product used here is based on the TIR CO absorption band. For this reason, they are not explicitly treated in the operational retrieval algorithm (Francis et al., 2026). We would like to note the study by Deeter et al. (2016), who found that average MOPITT CO bias w.r.t. two AERONET sites under high and low aerosol loads (in the Amazon basin) respectively were not significantly different. However, we must note here that this was for retrieval V6. From retrieval V8 to V9, changes in the cloud detection algorithm led to an increase to an increase in coverage over the Amazon region and particularly in conditions with high aerosol concentrations (Deeter et al., 2022).

On top of changing the light path, aerosols could influence retrievals if their impacts on micro-meteorological conditions are not well-represented in MERRA or ECMWF data, which could lead to errors in the temperature, water vapour and pressure profiles that the retrieval algorithms use.

While available validation studies do not indicate a substantial systematic aerosol-related bias in the products used here, we acknowledge that retrieval performance under extreme smoke conditions remains uncertain and an area of active research. We therefore added now more clearly acknowledge potential issues and give more background.

#### *Changes made to the manuscript:*

##### **Line 191 (Methods):**

The applied quality filter excludes observations under a range of potentially problematic cloud and aerosol conditions, although residual biases in dense smoke plumes cannot be completely ruled out.

##### **Line 511 (Discussion):**

Aerosol-induced  $X_{CO}$  retrieval biases are a potential concern in dense smoke plumes because aerosols modify the light path of photons which might affect retrieval sensitivity. However, aerosol scattering effects are explicitly accounted for in the SICOR retrieval and sensitivity tests presented in the algorithm theoretical baseline document that include cases with high aerosol loads, suggest the algorithm remains within 1% uncertainty (Landgraf et al., 2024). In addition, a validation campaign during a wildfire suggests that uncertainties remain within mission performance requirements even in optically thick smoke plumes (10% accuracy, 15% precision) (Rowe et al., 2022). Aerosol radiative effects in the 4.7  $\mu\text{m}$  thermal-infrared absorption band are weak (Eck et al., 1999) and therefore not explicitly accounted for in the retrieval (Francis et al., 2026). Deeter et al. (2016) tested for a shift of the mean in bias against two AERONET sites under high and low aerosol conditions, but did not find a significant shift. It must be noted that this was tested on version 6. Since then, specifically version 9, coverage increased, particularly under elevated aerosol conditions. Nonetheless, since aerosol properties, such as size and mixing state, are generally more complex than assumed in radiative transfer models (Guan et al., 2026a; Guan et al., 2026b), the effects of aerosols on retrievals remains a source of uncertainty.

*A second issue relates to the representativeness of the fire types sampled by the 125 ppb threshold. The 100/150 ppb sensitivity tests are a welcome addition and demonstrate that total fire season emissions are robust to the threshold choice (1.1 TgCO, or 2–3%).*

However, the authors’ own conclusion that “the primary mass constraint is driven by the heavier CO enhancements” (Sect. 2.5.4) suggests that low-intensity, diffuse fires contribute little to the mass constraint. This raises a question that: do the fires excluded by the 125 ppb threshold contribute a significant fraction of total emissions? If a substantial portion of emissions originates from low-intensity smoldering or understory fires that rarely produce XCO enhancements above 125 ppb, then the threshold would systematically miss these sources. A comparison of the FRP or active fire detection distributions for assimilated versus non-assimilated TROPOMI observations, or an evaluation of what fraction of GFED5.1/GFAS prior emissions is associated with low-intensity fire types, would help clarify whether the sampled fire types are representative of the full emission spectrum.

We thank the reviewer for raising this point. The key question is whether the 125 ppb threshold causes a substantial fraction of fire emissions to be missed by the inversion. We believe the threshold sensitivity experiment provides useful evidence. Lowering and increasing the threshold from 125 ppb to 100 and 150 ppb changes the posterior seasonal total by only 1.1 Tg CO (2–3%), despite the change in assimilated observations. This suggests that the region/season-total emissions are only weakly sensitive to the exact threshold choice.

We agree, however, that our statement “the primary mass constraint is driven by the stronger CO enhancements” warrants clarification. A weaker observational signature does not per definition mean that the corresponding emissions are ignored by the inversion. Emissions from low-intensity or diffuse fires are transported and mixed within the atmosphere and therefore contribute to the downwind X<sub>CO</sub> field sampled by the assimilated observations. Consequently, biases in such emissions still carry weight in the optimisation step through their contribution to the atmospheric CO signal. We have revised the text to more clearly explain this.

To further evaluate whether low-intensity fires could represent a substantial omitted source, Figure 1 shows the emission-rate distributions in GFED5.1 and GFAS v1.2 (as they are ingested in the transport model). Both inventories show a highly skewed distribution in which a large number of low-emission grid cells contribute only a small fraction of total emissions, while most emitted mass originates from a relatively small number of high-emission events. This behaviour is consistent with the threshold sensitivity experiment and suggests that observations excluded by the 125 ppb criterion are unlikely to account for a large fraction of the season-total emissions.

At the same time, we acknowledge an important limitation of the current framework. As discussed in Section 2.4, the inversion is not designed to independently constrain every fire at its exact location and time of occurrence. While the regional emission estimates appear robust to the threshold choice, the ability to resolve individual fire events remains limited, in particular when defined as zero-emission in the prior. In the manuscript we point to more flexible prior-error approaches, such as Yin et al. (2015), which may reduce spatio-temporal redistribution of missed or small fire emissions.

*Changes made to the manuscript:*

#### Line 251 (Methods):

These showed that the derived total fire emissions were barely affected by the exact threshold choice (a spread of 1.1 TgCO, or 2-3%, on Brazil fire season totals), **suggesting that the information content relevant to the regional emission budget is concentrated in observations associated with strong CO enhancements.**

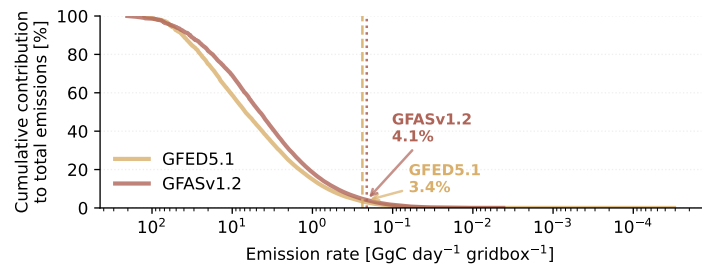


Figure 1: Cumulative contribution of all non-zero fire CO emission rates per day per 1° × 1° grid boxes from GFED5.1 (yellow) and GFASv1.2 (red) in the statevector domain during the simulation period. The contributions are relative to the simulation period/region total and sorted in descending order by emission rate strength. The vertical lines indicate the fraction of total emissions contributed by grid cells with emission rates below the median. The figure illustrates the strong skewness of fire emissions, with a large number of low-emission grid cells contributing only a small fraction of total fire CO emissions

## Reviewer I:

*The Introduction and Discussion sections could benefit from a broader overview of recent studies that use observational constraints to improve estimates of atmospheric composition, emissions, and associated climate impacts. In particular, several recent studies have highlighted the value of integrating satellite, ground-based, and multi-wavelength observations to better constrain pollutant emissions, carbon-related tracers, aerosol absorption properties, and radiative effects. Including a brief discussion of these developments would help place the present work into a wider context of observation-constrained atmospheric and carbon-cycle research.*

*Suggested references:*

Guan, L., Cohen, J. B., Wang, S., Tiwari, P., Liu, Z., & Qin, K. (2026). Improving aerosol absorption estimates via size-resolved constraints based on AERONET and in situ measurements. *Geophysical Research Letters*, 53(5), e2025GL117418.

Guan, L., Cohen, J. B., Wang, S., Tiwari, P., Liu, Z., Li, Z., & Qin, K. (2026). In-tandem multi-waveband particulate absorption and size observations yield substantial changes in radiative forcing over industrial Central China. *Atmospheric Chemistry and Physics*, 26(4), 3107–3123.

Wang, S., Cohen, J. B., Guan, L., Lu, L., Tiwari, P., & Qin, K. (2025). Observationally constrained global NO<sub>x</sub> and CO emissions variability reveals sources which contribute significantly to CO<sub>2</sub> emissions. *npj Climate and Atmospheric Science*, 8(1), 87.

We thank the reviewer for this suggestion. We agree that the broader field of observation-constrained atmospheric composition research has expanded considerably in recent years, including applications that use satellite and ground-based observations to improve estimates of emissions, atmospheric composition, aerosol properties, radiative forcing, and carbon-cycle processes. While the focus of the present study remains CO fire emissions, we have expanded the discussion section to place our work within this emerging class of observation-constrained approaches. In particular, we now discuss recent studies that use observational constraints to improve estimates of NO<sub>x</sub> and CO emissions (Wang et al., 2025), aerosol absorption and radiative forcing (Guan et al., 2026a; Guan et al., 2026b), and the potential for future multi-tracer, multi-instrument inversion systems.

*Changes made to the manuscript:*

### Line 511 (Discussion):

Aerosol-induced  $X_{CO}$  retrieval biases are a potential concern in dense smoke plumes because aerosols modify the light path of photons which might affect retrieval sensitivity. However, aerosol scattering effects are explicitly accounted for in the SICOR retrieval and sensitivity tests presented in the algorithm theoretical baseline document that include cases with high aerosol loads, suggest the algorithm remains within 1% uncertainty (Landgraf et al., 2024). In addition, a validation campaign during a wildfire suggests that uncertainties remain within mission performance requirements even in optically thick smoke plumes (10% accuracy, 15% precision) (Rowe et al., 2022). Aerosol radiative effects in the 4.7  $\mu\text{m}$  thermal-infrared absorption band are weak (Eck et al., 1999) and therefore not explicitly accounted for in the retrieval (Francis et al., 2026). Deeter et al. (2016) tested for a shift of the mean in bias against two AERONET sites under high and low aerosol conditions, but did not find a significant shift. This finding, however, related to version 6 of the MOPITT algorithm. Especially since version 9 coverage increased, particularly under elevated aerosol conditions. **Nonetheless, since aerosol properties, such as size and mixing state, are generally more complex than assumed in radiative transfer models (Guan et al., 2026a; Guan et al., 2026b), the effects of aerosols on retrievals remains a source of uncertainty.**

### Line 525 (Discussion):

With continuing advances in computational capacity, increasing availability of large atmospheric datasets, as well as capacity to efficiently analyse these, multi-tracer, multi-instrument approaches are on the rise (e.g. Wang et al., 2025). From a fire-emissions inversion perspective, exploration of joint observation constraints from CO, CO<sub>2</sub>, NO<sub>2</sub>, aerosols, HCHO, and CH<sub>4</sub>, may not just benefit total fire emission quantification, but also help to partition in terms of fire type or burning phase, as well as the ability to separate source-background signals.

## References

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