

Responses to Reviewers' Comments

We thank the reviewers for careful reading and valuable comments. Here, we use black for the reviewer's comments, blue for our responses, and italic for the manuscript, where the changes are highlighted in red.

Reviewer #1

General Comments:

This manuscript presents a comprehensive study assimilating satellite observations of six key chemical species into the MIROC-Chem model to simultaneously optimize atmospheric concentrations and surface emissions. The assimilated results are extensively validated against the recent NASA ASIA-AQ airborne observations, demonstrating a substantial reduction in model biases. The optimized emissions reveal significant discrepancies in current bottom-up inventories for anthropogenic sources, biogenic sources, and biomass burning. Furthermore, the study provides a detailed source-receptor analysis quantifying the relative contributions of local emissions versus long-range transport to air quality in various Asian cities. Overall, the methodology is solid and robust, and the conclusions are well-supported by the analysis. The manuscript is clearly written and well-structured. I recommend publication after the following specific comments and clarifications are addressed.

Specific Comments:

1. The current inversion framework attributes all simulated concentration biases to errors in surface emissions. How can we be certain that these biases do not stem from inherent deficiencies in the model's physical or chemical mechanisms (e.g., boundary layer mixing, deposition schemes, or chemical reaction rates)? I suggest adding a brief discussion on the potential uncertainties introduced by model structural errors and how they might alias into the optimized top-down emissions.

Thank you for the comment. We have revised the paper to clarify the following points.

1. In our data assimilation system, the state vector includes both chemical concentrations and emissions. Model-observation discrepancies are therefore not attributed exclusively to emission errors. The analysis increments are distributed among these state variables according to the observation and background error covariances.
2. This approach differs from inversion approaches in which emissions are the only optimized state variables and model-observation discrepancies are directly mapped onto emission adjustments.
3. The background error covariances evolve with the atmospheric state. Therefore, the relative adjustments made to concentration and emission vary in space and time. The cross-variable covariance structures are defined separately for different emission species.

To explain them and the influence of model structural errors, we have added the following discussion to Section 4.

These top-down estimates are subject to several sources of uncertainty. Forward model errors in chemistry, boundary-layer mixing, deposition, and transport can propagate into the inferred emissions. For example, Miyazaki et al. (2020a) showed that the sensitivity of surface O_3 and NO_2 to NO_x emissions differed by up to a factor of two among models, reflecting differences in the representation of fast chemical and dynamical processes. In our framework, chemical concentrations and emissions are optimized simultaneously, which allows part of the model-observation mismatch arising from non-emission errors to be absorbed by concentration adjustments rather than entirely by emission adjustments

(Miyazaki and Eskes, 2013). Multi-species constraints further matter; assimilating species other than NO₂ can alter regional monthly mean NO_x emissions by up to 70% (Miyazaki et al., 2017). Additional uncertainties arise from the coarse horizontal resolution, which cannot resolve urban plumes and their nonlinear chemistry (Valin et al., 2011; Sekiya et al., 2021), biases in the assimilated retrievals, and uncertainties in the prior inventories. The combined influence of retrieval biases and prior inventories is illustrated by the two CO configurations in this study (Sections 2.1 and 3.1), which yield biomass burning CO estimates over Thailand differing by roughly a factor of two (Section 4.2).

2. Please elaborate in the methodology section on how the data assimilation system handles this strong non-linear chemical feedback (for example, the strong non-linearity in ozone chemistry). Does this non-linearity pose a risk for optimizing NO_x and VOCs emissions during the assimilation process?

Chemical nonlinearity may introduce uncertainty into the optimization of NO_x and VOC emissions. In our system, the background error covariance is estimated every two hours from an ensemble propagated with the full nonlinear model. This allows the relationships between emissions and chemical concentrations to evolve with the local chemical regime. We have added a brief explanation of this treatment to Section 2.1.

In Section 2.1:

The TCR-3 product, generated using the MOMO-Chem framework, is used in this study (Miyazaki et al., 2020a, 2020b). It employs an ensemble-based data assimilation system based on the Local Ensemble Transform Kalman Filter (LETKF) (Hunt et al., 2007). The state vector includes various chemical concentrations as well as surface emissions of CO, NO_x, SO₂, and isoprene and lightning production of NO_x, with background error covariance linking observed atmospheric concentrations to emission adjustments for each model grid cell. The background error covariance is estimated at each analysis step (every two hours) from ensemble forecasts propagated with the nonlinear chemical transport model, allowing the flow-dependent covariance to evolve with the chemical state. The short assimilation cycle supports gradual adjustments to the analysis and allows observation-model differences to be corrected iteratively through data assimilation cycles. These features allow the analysis to adapt to spatially and temporally varying chemical regimes, including transitions between NO_x-saturated and NO_x-limited conditions.

3. The study assimilates six gas-phase species but does not mention aerosols (e.g., PM_{2.5} or AOD). Asia, particularly India and Thailand during the fire season, experiences extremely high aerosol loadings. Heavy aerosol plumes can significantly alter photolysis rates. I recommend evaluating, or at least discussing, the potential errors introduced into the gas-phase photochemistry and the subsequent emission inversions due to the lack of aerosol assimilation (or unconstrained AOD).

We assimilated VIIRS AOD to optimize primary aerosol emissions (Section 2.1). The benefits of the simultaneous aerosol and trace gas assimilation have been demonstrated in detail by Sekiya et al. (2026). This evaluation has been extended to a longer period, including the ASIA-AQ campaign (manuscript in preparation). Because the present study focuses primarily on trace gas analyses and emission estimates, we do not include a separate evaluation of the aerosol fields here. However, we agree that aerosol constraints are important for representing aerosol-photolysis interactions and their influence on gas-phase chemistry. We have therefore emphasized this point more clearly in the revised manuscript as follows:

In Section 4:

Along with the trace gas observations, we assimilate VIIRS AOD observations to optimize primary aerosol emissions. This is expected to improve the representation of aerosol loading and its influence on gas-phase photochemistry, particularly under conditions strongly affected by biomass burning. Sekiya et al. (2026) have confirmed improvements in the aerosol fields across a range of conditions, and a detailed evaluation over an extended period, including the ASIA-AQ campaign, will be presented in a separate study.

4. How is the injection height of Southeast Asian wildfires parameterized in the model? Since the vertical distribution of emissions is critical for simulating long-range transport, did the ASIA-AQ airborne observations provide any constraints or validation for the modeled plume injection heights? Please provide more information on this in the text.

In our configuration, biomass burning emissions are released at the surface without a plume-rise parameterization. The DC-8 profiles provide valuable information on the vertical profiles of fire-emitted species, but interpretation of elevated layers requires separating the effects of local plume injection from long range transport. To discuss this, we have revised the relevant sentence as follows:

In Section 3.1:

This trade-off (better free-tropospheric performance at the cost of surface overestimation) is particularly evident in Northern Thailand and may suggest that the model places too large a fraction of these emissions near the surface, or that vertical mixing in the model does not efficiently redistribute them aloft. This also points to uncertainties in the vertical distribution of biomass burning emissions (Pan et al., 2026). Developing an appropriate plume-rise parameterization and evaluating it against aircraft observations will be an important direction for future work, which will require distinguishing locally injected plumes from elevated layers associated with long-range transport.

5. Line 376-385: The model scales down biogenic isoprene emissions by a striking 52%. Is this massive reduction primarily due to an overestimation of basal emission factors in the prior model (e.g., MEGAN), or is it caused by inadequate parameterization of the vegetation's response to meteorology (e.g., drought stress) for specific Asian land cover types? I suggest providing a more in-depth mechanistic explanation, possibly by linking the results to meteorological fields (e.g., temperature, PAR) and land cover data.

The large emission reduction may reflect uncertainties in both the bottom-up inventory and the model chemistry. Its magnitude is broadly consistent with previous studies reporting that MEGAN overestimates isoprene over Southeast Asian tropical forests by factors of 2–4 (Langford et al., 2010; Stavrou et al., 2014). A recent model intercomparison (Do et al., 2025) also shows a large spread in Southeast Asian isoprene emissions, primarily associated with differences in plant functional types and assigned emission factors. In addition, uncertainties in isoprene oxidation chemistry can largely affect simulated VOC concentrations (e.g., Yang et al., 2023) and therefore the inferred emissions. A quantitative attribution among these factors would require dedicated sensitivity experiments in future studies. We have revised the discussion to clarify these sources of uncertainty.

In Section 4.1:

~The adjustments underscore the substantial uncertainties in bottom-up isoprene inventories over Asian tropical regions, with important implications for O₃ production, OH budgets, and secondary organic aerosol formation (Yoon et al., 2025). A model intercomparison similarly found Southeast Asian isoprene emissions differed by approximately a factor of six across models (14.7–87.8 TgC yr⁻¹), driven mainly by

differences in the representation of plant functional types and their assigned emission factors (Do et al., 2025). At the same time, the top-down isoprene estimates remain subject to considerable uncertainty, because they depend on the representation of isoprene chemistry and the broader chemical environment within the model (Choi et al., 2025; Wells et al., 2020; Yang et al., 2023). For example, Yang et al. (2023) found that replacing the default GEOS-Chem chemistry with a newly developed isoprene oxidation algorithm increased simulated isoprene column concentrations by 38% over the southeastern United States. This result illustrates how the choice of chemical mechanism alone can affect simulated concentrations and then the inferred isoprene emissions. In addition, potential biases in the assimilated TROPOMI CH₂O retrievals may influence the optimized emissions and require further investigation (Oomen et al., 2024).

6. Section 5: The analysis of local and transboundary transport based on regional emission perturbation studies is highly appreciated, as it requires significant computational effort. However, the manuscript does not seem to address the non-linear interactions between emission changes from different regions. For instance, in the pie charts (e.g., Figs. 13 and 14), the sum of individual regional contributions theoretically shouldn't perfectly equal 100% due to chemical non-linearities. Are these interaction terms lumped into the "rest of SE Asia" categories? Please clarify how these non-linear interactions are accounted for in the fractional attributions.

We thank the reviewer for this point. Because of chemical nonlinearity, the sum of the responses to individual regional perturbations does not necessarily equal the response to a simultaneous perturbation of all regions. In the pie charts, the fractional contributions are calculated by normalizing each regional response by the sum of the responses from all individual source regions. The “rest of SE Asia” category represents a distinct source region within the Asia domain. We have clarified this treatment in the revised manuscript.

We have clarified the nonlinearity in the revised paper, based on the 20% and 100% (zero-out) perturbation experiments (Fig. 13 and Fig. S9). The main text focuses on the 20% reductions because modest emission cuts are more relevant to near-future mitigation scenarios, consistent with the framing adopted in recent source-attribution studies (Choi et al., 2024; Gu et al., 2023; Nawaz et al., 2023). The zero-out experiments are retained to demonstrate how the inferred regional influences change under much larger perturbations.

In Section 5,

Figure 13 shows the daily surface O₃ response in five receptor cities (Seoul, Chiang Mai, Taipei, Manila, and Bangkok) to 20% emission reductions applied separately to each source region within the Asia domain (Fig. S8), representing a near-future mitigation scenario. The fractional contributions in the pie charts are calculated by normalizing each regional response by the sum of the responses from all individual regional perturbations. The fractions therefore sum to 100% by construction. Because of chemical nonlinearity, however, the sum of the individual responses does not necessarily equal the response to a simultaneous reduction across all regions. To examine this nonlinearity, the corresponding 100% emission reduction (zero-out) experiments are shown in Fig. S9. A comparison between the two experiments reveals substantial nonlinearity in the O₃ response, particularly over Seoul and Taipei, where the 100% response is not proportional to the 20% response. For example, over Seoul on February 11, removing 100% of mainland China emissions produces an O₃ response of ~–30 ppbv, whereas a 20% reduction yields only ~–2 ppbv, far less than one-fifth of the zero-out response, reflecting NO_x-saturated photochemical conditions and the nonlinear sensitivity of O₃ to precursor emissions. In general, foreign emission contributions appear larger in the 100% sensitivity simulation, showing that the 20% reduction

results presented here provide a conservative and policy-relevant estimate of transboundary influences under realistic mitigation scenarios.

Technical Corrections:

7. Please replace "country" with "country/region" where appropriate (for example, in the colorbar of Fig. 2h or similar figures and texts) to maintain geopolitical neutrality and adhere to standard scientific publication guidelines regarding disputed territories.

We thank the reviewer for this suggestion. We have replaced "country" with "region" where appropriate: the colorbar of Fig. 2h and the corresponding label in Fig. 11.

Reviewer #2

Choi et al. perform a comprehensive study aiming to assess air pollution in Asia backed by the NASA ASIA-AQ airborne mission campaign measurements of ozone, NO_x, CO and CH₂O. To this purpose, they apply a multi-constituent satellite data assimilation approach using MLS profile retrievals of ozone, CO and HNO₃, NO₂, CH₂O and SO₂ columns from TROPOMI, and VIIRS AOD products. The baseline framework assimilates CO column data from MOPITT, whereas a sensitivity experiment using TROPOMI CO columns is also conducted and evaluated against ASIA-AQ data. The chemical assimilation method was previously developed and used in a wide range of studies (Miyazaki et al. <https://doi.org/10.5194/essd-12-2223-2020>). The current Tropospheric Chemical Reanalysis (TCR-3) includes only minor updates relative to the previous versions (TCR-2). After assimilation, the agreement with the airborne data was generally improved, as a consequence of moderate emission increases for anthropogenic NO_x and CO, a strong decrease for isoprene, and a pronounced increase in fire emission estimates in Thailand. The analysis reveals that meteorology governs the transboundary pollution in this region. The study is useful for the community. The manuscript is clear, well written and the results look correct. I recommend publication after addressing the comments below.

- The reasons for the large differences (Figure 5) between the two ASIA_AQ ozone measurements acquired with different techniques (in situ cavity-enhanced UV absorption of ozone and chemiluminescence) should be discussed. In Taiwan, for example, if NO_x emissions were increased to match the ASIA-AQ NO_x observations, this would lead to ozone levels much higher than the measurements. This does not seem consistent. Please explain.

We thank the reviewer for highlighting this issue. The differences between the two ozone measurements exceed their reported uncertainties, but their cause has not been conclusively resolved by the measurement teams. We are not in a position to determine which dataset provides the more accurate reference and have retained both in Fig. 5 to represent this observational uncertainty. This uncertainty affects the ozone evaluation across the campaign domain, not only over Taiwan. Relative to NO_xO₃, the assimilation reduces the negative bias (NMB from -17% to -14%), whereas relative to ROZE, it increases the positive bias (+7% to +12%), although R improves against both (Table 2). To clarify these implications for the O₃ evaluation, we have added the following sentences to section 3.3.

Whether the assimilation reduces the O₃ bias depends on the reference dataset, both across the campaign domain and within individual regions (Fig. 5), whereas the improvement in R is consistent across the two instruments. This unresolved discrepancy between the measurements introduces uncertainty into the evaluation of the O₃ response to the optimized precursor emissions.

Regarding the consistency between NO_x and O₃ over Taiwan, the assimilation increases Taiwan NO_x emissions by only 14%, and the simulated NO_x remains substantially lower than observed. This discrepancy is partly because urban NO_x plumes are not resolved at the model's coarse resolution. However, a further NO_x increase would not necessarily translate into proportionally higher O₃, particularly under NO_x-saturated conditions (e.g., Taipei in Fig 13. b), where NO titration can offset ozone production.

- The use of TROPOMI CO in TCR-3 significantly improves the comparison with ASIA-AQ measurements relative to MOPITT, as seen in Fig. 5. The difference between the two assimilations is quite large but is not adequately explained. Are the TROPOMI columns higher than MOPITT in this region? Comparisons between TROPOMI and MOPITT (Martinez-Alonso et al. <https://amt.copernicus.org/articles/13/4841/2020/amt-13-4841-2020.pdf>) indicated an excellent

agreement over land, with TROPOMI only slightly lower than MOPITT. Can you provide additional analysis and independent measurements to explain the large difference? On the other hand, the TROPOMI TCR-3 emissions are much lower than the MOPITT TCR-3 in Figure 8f over Thailand, but this seems to be inconsistent with Fig. 5l. Please explain. Could you provide additional plots as Figure 6 (b,e) but for TROPOMI TCR-3?

We first note that the two CO assimilation runs differ in both the prior emissions and the assimilated observations. The standard TCR-3 (MOPITT) uses HTAPv3 and GFASv1.2 as priors and assimilates MOPITT CO, whereas TCR-3 (TROPOMI) uses CEDS and GFED4 and assimilates TROPOMI CO. Their difference therefore cannot be attributed to the CO retrieval alone, as stated in Section 4.2. These two configurations were developed for different purposes (long-term reanalysis vs short-term event analysis), and therefore do not provide a controlled comparison of retrieval impacts. Experiments designed to isolate the retrieval effects will be presented in the forthcoming TCR-3 description paper, although for a different period.

The agreement between MOPITT and TROPOMI columns reported by Martínez-Alonso et al. (2020) also does not by itself imply agreement in the inferred emissions. Assimilation distributes each observation-model departure onto concentrations and emissions according to the background and observation error covariances. The resulting increments also depend on the observation errors, the vertical sensitivity represented by the averaging kernels, and the sampling and spatial resolution ($\sim 22 \times 22$ km for MOPITT and $\sim 7 \times 5.5$ km for TROPOMI), all of which differ between the two instruments. Thus, similar columns can lead to different emission increments.

The apparent inconsistency between Figs. 8f and 5l also reflects the different quantities being compared. Figure 8 compares optimized anthropogenic CO emissions, whereas the CO profiles in Fig. 5l are strongly influenced by biomass-burning emissions. The TROPOMI assimilation provides particularly strong constraints on the fire-related CO enhancements during this period. Therefore, the relative differences between the TROPOMI- and MOPITT-based assimilations do not need to be consistent between Fig. 5l and Fig. 8.

The requested fields analogous to Fig. 6 (b,e) are provided in Fig. S6 for both configurations.

- The large inferred decrease of isoprene emissions in South East Asia is well supported by ASIA-AQ CH₂O measurements, and an even stronger decrease would be required, especially in South Korea and Thailand (Fig. 5o,p). However, recent top-down isoprene emissions based on TROPOMI CH₂O data (Sfendla et al. <https://doi.org/10.5194/acp-26-733-2026>, Li et al. <https://doi.org/10.1021/acs.est.5c12927>) and CrIS isoprene (Li et al. <https://doi.org/10.5194/essd-17-7035-2025>) found generally higher annual emission fluxes than the a priori. Could you compare with your results for the specific months? It is also recommended to evaluate against other bottom-up estimates, e.g. with CAMS-GLOB-BIO (Sindelarova et al. <https://doi.org/10.5194/essd-14-251-2022>).

We thank the reviewer for these references. We would first note that these studies report annual-mean fluxes over broader or different domains, whereas our estimates cover February–March over Southeast Asia. Therefore, they do not provide a direct comparison for the same region and period. Nevertheless, accounting for regional differences and retrieval reliability largely explains the apparent disagreement in the direction of the emission adjustments:

- **Li et al. (CrIS isoprene, ESSD 2025):** In their Fig. 1, the posterior is lower than the prior over Southeast Asia, consistent with our result. Their higher-than-prior fluxes occur over Northeast Asia. However, CrIS isoprene there falls below the detection limit, which limits the information gained from the inversion in that region (Choi et al., 2025). This is especially the case in the boreal cold season of our interest (Feb–Mar 2024). The CrIS result is therefore consistent with ours where the observations are reliable (Southeast Asia) and uninformative where they diverge (Northeast Asia).
- **Li et al. (TROPOMI CH₂O, EST 2026):** This study reaches the same conclusion for our region: their posterior-versus-prior isoprene comparison over Southeast Asia (Fig. S18b) and the corresponding simulated concentrations (Fig. S19) also show a posterior below the prior, consistent with our inferred decrease.
- **Sfendla et al. (TROPOMI HCHO, ACP 2026):** Their optimized HCHO columns exceed the a priori columns over the region (e.g., Fig. 1 for DJF 2021), consistent with higher-than-prior annual emissions (Fig. 2), but the comparison is not directly applicable: they use a different forward model and, importantly, a different a-priori biogenic inventory (MEGAN-MOHYCAN) and anthropogenic/fire inventories (CAMSGLOBANT, QFED). Because a top-down increment is defined relative to the prior, a different prior can yield an opposite-signed adjustment.

We additionally compare our estimates with CAMS-GLOB-BIOv4.0 for February–March 2024. Over the Asia domain, CAMS-GLOB-BIOv4.0 yields 13.9 Tg of isoprene, compared with 9.3 Tg in TCR-3. Over Southeast Asia, the corresponding values are 8.8 and 5.3 Tg. The TCR-3 isoprene emissions are 30–40% lower than the CAMS-GLOB-BIOv4.0.

We have revised our Section 4.

Recent top-down estimates using TROPOMI CH₂O and CrIS isoprene similarly indicate lower posterior than prior emissions over Southeast Asia (Li et al., 2025, 2026), consistent with our adjustments, although the periods differ. TCR-3 isoprene emissions (9.3 Tg) are also 33% lower than CAMS-GLOB-BIOv4.0 (13.9 Tg) (Sindelarova et al., 2022) over the domain for the same period.

- The coarse resolution of the assimilation is a limitation when we address short-lived species. As seen in Figure 5, the NO_x emission increase in TCR-3 is relatively small and the model remains too low compared to the ASIA-AQ measurements. What is the posterior model bias with respect to the satellite NO₂ used in the assimilation? Do you have the possibility to conduct a control run at finer resolution? Could you add a comparison with available Pandora measurements in your target box?

We thank the reviewer for this comment. Figure S2 (new) compares the CTL and TCR-3 tropospheric NO₂ columns with the assimilated TROPOMI NO₂ columns for February–March 2024. The assimilation substantially improves the spatial agreement. CTL overestimates NO₂ over Northeast Asia (North China Plain, South Korea), Bangkok, and India, and underestimates it over mainland Southeast Asia. Most of these biases are substantially reduced in TCR-3. Assimilation increases R from 0.89 to 0.96 and reduces the RMSE from 8.3×10^{14} to 3.1×10^{14} molec cm⁻². The domain-mean NMB nevertheless changes from +1.4% to -8.4%, because the regional overestimates and underestimates in CTL largely cancel in the domain mean. The directions of these adjustments are consistent with the DC-8 comparison: the near-surface NO_x bias over South Korea shifts from positive to negative, while NO_x increases over the Philippines and Taiwan (Section 3.4).

Improved agreement with satellite columns does not necessarily translate into improved agreement with DC-8 measurements. Assimilation constrains the surface emissions at the model resolution but does not directly constrain the vertical distribution of NO₂ or the representation of localized plumes.

Following the reviewer's suggestion, we also evaluated the model against Pandora tropospheric NO₂ columns (Fig. R1). During the campaign period, nine Pandora sites within our target regions had valid data: eight in South Korea and one in Bangkok. Both runs underestimate the observed columns, with a larger negative bias in TCR-3 (NMB -57%) than in CTL (-35%). This is consistent with the DC-8 evaluation. These results further demonstrate that improved agreement with the assimilated satellite columns can coexist with larger biases against local observations. The coarse horizontal resolution is an important limitation for representing short-lived species such as NO₂, although differences in vertical sensitivity and sampling may also contribute to these discrepancies. A finer-resolution control run is beyond the scope of this study. Previous work showed that assimilation at 0.56° reduced biases against independent surface NO₂ observations more effectively than at 1.1° or 2.8° in many cases (Sekiya et al., 2021). We have revised Section 3.4 to clarify these limitations.

Underestimation of boundary layer NO_x has been commonly reported by previous studies (Miyazaki et al., 2012; Skoulidou et al., 2021). Relative to the assimilated TROPOMI NO₂ columns, the assimilation removes most of the regional biases in CTL and reduces the RMSE from 8.3×10^{14} to 3.1×10^{14} molec cm⁻² (Fig. S2). However, improved agreement with satellite columns does not consistently translate into improved agreement with DC-8 measurements and Pandora tropospheric NO₂ column measurements (figure not shown). At the model resolution, satellite columns neither constrain the vertical distribution of NO₂ nor resolve localized NO_x plumes and their non-linear chemistry, highlighting the need for higher model resolution (Valin et al., 2011; Sekiya et al., 2021). Evaluation against NO₂ measurements from NOxO3 shows a similar pattern (not shown). CTL shows a near-neutral domain-wide bias (+2% NMB) with R of 0.68, while TCR-3 introduces a negative bias (-19% NMB) with R reducing to 0.62, consistent with the NO_x results. In contrast to NO_x, whose RMSE increases slightly with assimilation (Table 2), RMSE for NO₂ improves from 3.0 to 2.6 ppbv.

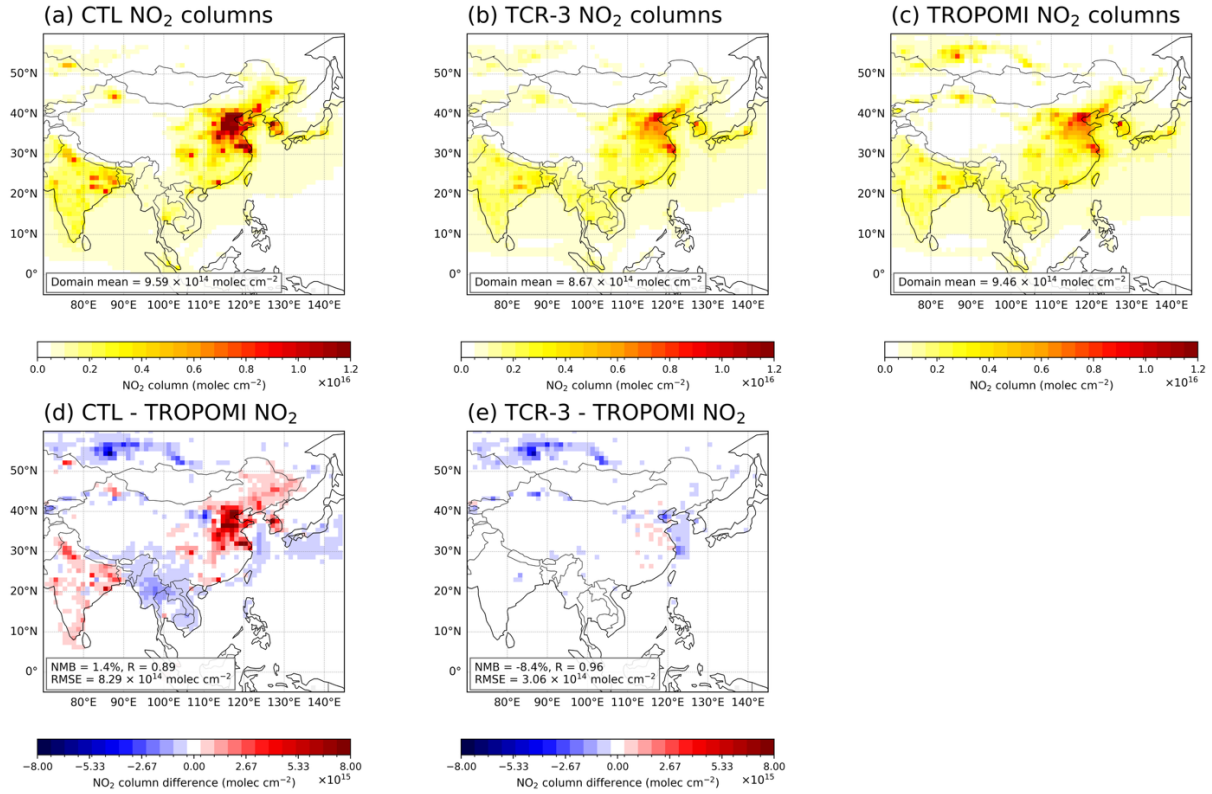


Figure S2. Tropospheric NO_2 columns averaged over February-March 2024 from (a) CTL, (b) TCR-3, and (c) TROPOMI, and differences relative to TROPOMI for (d) CTL and (e) TCR-3. All fields are shown on the TCR-3 grid. Model columns are sampled at the TROPOMI overpass times and locations. Domain means are shown in panels (a-c), and NMB, R, and RMSE relative to TROPOMI are shown in panels (d-e).

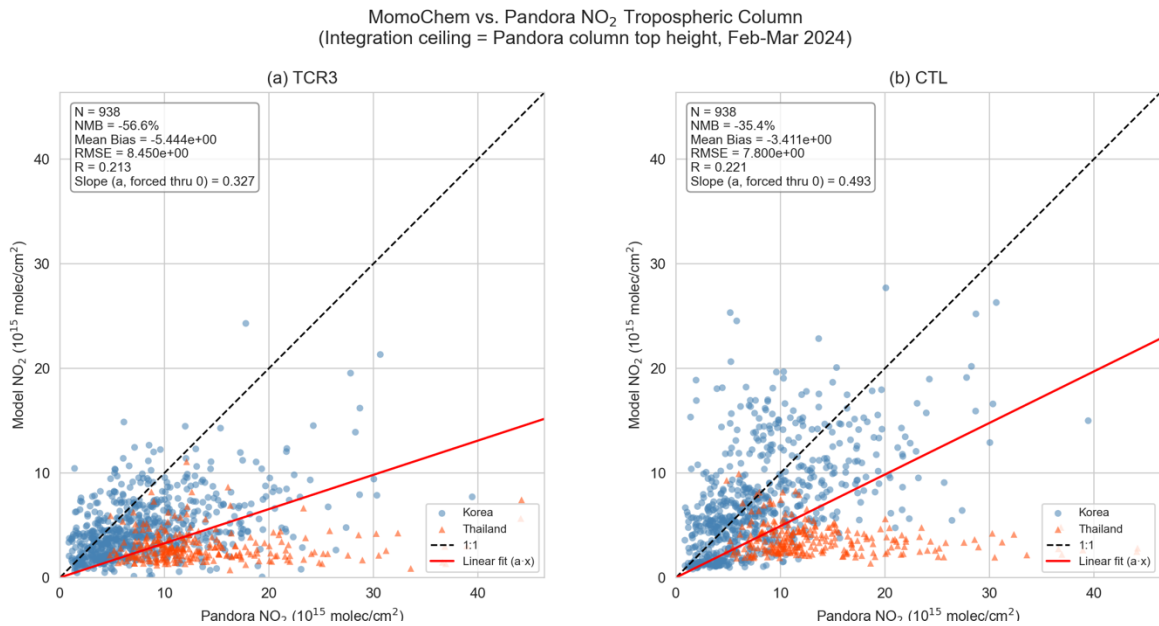


Figure R1. Model versus Pandora tropospheric NO₂ vertical columns during ASIA-AQ (February–March 2024).

Scatter of MOMO-Chem NO₂ tropospheric columns against co-located Pandora MAX-DOAS retrievals (PGN product rnvh3) for (a) the TCR-3 assimilation run and (b) the control (CTL) run. Pandora measurements from nine sites (eight in South Korea and Bangkok in Thailand) are grouped onto the four model grid cells they fall within, and each point is a model cell $\times$ 2 hourly match ($N = 938$), colored by region. The five Seoul area sites (Seoul, Seoul-SNU, Incheon-ESC, Suwon-USW, and Yongin) fall in one cell, Busan and Ulsan share another, and Seosan and Bangkok each occupy their own; the Pandora retrievals within a cell are compared against that cell's single model column. Model columns are formed by integrating the model NO₂ profile from the surface to each observation's Pandora column top height (~ 3 to 4 km). The dashed line is 1:1 and the red line is a least square fit through the origin (slope a); N , normalized mean bias (NMB), mean bias, RMSE, correlation (R), and slope are inset. Both runs underestimate the observations, with TCR-3 lower than CTL (NMB -56.6% vs -35.4% ; slope 0.33 vs 0.49), indicating that assimilation of coarse resolution satellite NO₂ does not fully close the gap for this short-lived species.

New references cited in this response

Choi, J., Henze, D. K., Nawaz, M. O., and Malley, C. S.: Source Attribution of Health Burdens From Ambient PM_{2.5}, O₃, and NO₂ Exposure for Assessment of South Korean National Emission Control Scenarios by 2050, *GeoHealth*, 8, e2024GH001042, <https://doi.org/10.1029/2024GH001042>, 2024.

Do, N. T. N., Sudo, K., Ito, A., Emmons, L. K., Naik, V., Tsigaridis, K., Seland, Ø., Folberth, G. A., and Kelley, D. I.: Historical trends and controlling factors of isoprene emissions in CMIP6 Earth system models, *Geoscientific Model Development*, 18, 2079–2109, <https://doi.org/10.5194/gmd-18-2079-2025>, 2025.

Gu, Y., Henze, D. K., Nawaz, M. O., and Wagner, U. J.: Response of the ozone-related health burden in Europe to changes in local anthropogenic emissions of ozone precursors, *Environ. Res. Lett.*, 18, 114034, <https://doi.org/10.1088/1748-9326/ad0167>, 2023.

Li, H., Ciais, P., Kumar, P., Hauglustaine, D. A., Chevallier, F., Broquet, G., Millet, D. B., Wells, K. C., Lian, J., and Zheng, B.: Global biogenic isoprene emissions 2013–2020 inferred from satellite isoprene observations, *Earth System Science Data*, 17, 7035–7054, <https://doi.org/10.5194/essd-17-7035-2025>, 2025.

Li, H., Ciais, P., Kumar, P., Broquet, G., Chevallier, F., Hauglustaine, D. A., Millet, D. B., Wells, K. C., Lian, J., Bourtsoukidis, E., Zhang, K., and Zheng, B.: Contrasting Biogenic Isoprene Emission Responses to La Niña and El Niño Driven by Temperature: Insights from HCHO-Based Global Inversion, *Environmental Science & Technology*, 60, 3403–3415, <https://doi.org/10.1021/acs.est.5c12927>, 2026.

Miyazaki, K. and Eskes, H.: Constraints on surface NO_x emissions by assimilating satellite observations of multiple species, <https://doi.org/10.1002/grl.50894>, 2013.

Miyazaki, K., Eskes, H., Sudo, K., Boersma, K. F., Bowman, K., and Kanaya, Y.: Decadal changes in global surface NO_x emissions from multi-constituent satellite data assimilation, *Atmospheric Chemistry and Physics*, 17, 807–837, <https://doi.org/10.5194/acp-17-807-2017>, 2017.

Nawaz, M. O., Henze, D. K., Anenberg, S. C., Braun, C., Miller, J., and Pronk, E.: A Source Apportionment and Emission Scenario Assessment of PM_{2.5}- and O₃-Related Health Impacts in G20 Countries, *Geohealth*, 7, e2022GH000713, <https://doi.org/10.1029/2022GH000713>, 2023.

Sekiya, T., Miyazaki, K., Eskes, H., Rijdsdijk, P., Sudo, K., and Kanaya, Y.: Global aerosol composition constraints from simultaneous data assimilation of satellite AOD and trace gas observations, *EGUsphere*, 1–49, <https://doi.org/10.5194/egusphere-2026-1681>, 2026.

Sindelarova, K., Markova, J., Simpson, D., Huszar, P., Karlicky, J., Darras, S., and Granier, C.: High-resolution biogenic global emission inventory for the time period 2000–2019 for air quality modelling, *Earth System Science Data*, 14, 251–270, <https://doi.org/10.5194/essd-14-251-2022>, 2022.