

Reply to Comments by Anonymous Referee #2 on:

Global aerosol composition constraints from simultaneous data assimilation of satellite AOD and trace gas observations

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We would like to thank anonymous referee #2 for their careful reading of our manuscript and for providing valuable comments, which have greatly improved the manuscript. We have revised the manuscript accordingly, and the main changes are summarized below:

- 1) Add a schematic matrix that state variables whether these updates occur directly during the analysis in Figure 1 to clarify the pathways for updates of concentrations and emissions analyses.
- 2) Quantify impacts of direct concentration modifications and emission optimizations on aerosol concentration analyses using the additional data assimilation calculation without emission optimization (Fig. 2).
- 3) Add the validations using SO₂, NO₂, and ozone in-situ observation networks to Table 3 and Figs S4-6.

Individual comments (in black) and point-by-point replies (in blue) are listed below. Revised texts (*italicized font*) from the updated manuscript are in quotes below.

This manuscript presents a significant extension of the MOMO-Chem data assimilation framework by developing a coupled gas–aerosol assimilation system that simultaneously optimizes aerosol concentrations and emissions using satellite observations of aerosol optical depth (AOD) together with multiple trace-gas species (NO₂, SO₂, CO, and HNO₃). The authors demonstrate that combining aerosol and gas observations provides improved analyses of sulfate, nitrate, ammonium, and PM_{2.5} compared to assimilating either observation type alone, and they perform a comprehensive evaluation using independent aircraft and surface observations across multiple regions. The manuscript is well organized, the experimental design is carefully executed, and the validation is thorough. The study represents a valuable contribution to the fields of atmospheric chemistry and data assimilation by advancing the use of multi-species observations to constrain aerosol composition and precursor emissions. Overall, I find the paper to be scientifically sound and

suitable for publication. I recommend publication after the authors have addressed the following comments:

We appreciate the careful reviews again and positive comments.

Incomplete description of Cross-species covariance and variable localization in methodology

The manuscript methodology section introduces variable localization as the mechanism used to suppress spurious cross-species ensemble correlations, but its implementation is described only briefly. Section 2.1.2 states that variable localization is applied to eliminate spurious correlations and that the system prioritizes robustness of individual emission estimates, yet it does not specify which cross-species covariance blocks are retained or suppressed, nor the physical rationale underlying these choices.

Thank you for helpful comment. We added a schematic matrix that state variables whether these updates occur directly during the analysis in Figure 1. In addition, we clarified the reason underlying these choices:

(p. 6, l. 158–170)

“Specifically, as summarized in Fig. 1, ..., AOD with the observed Ångström exponent (α) of > 0.5 for primary carbonaceous aerosol emissions, and AOD with α of ≤ 0.5 for soil dust aerosol emissions. The Ångström exponent-based data screening can select dust-dominated coarse-mode AOD data, while we remain the assumption of size distribution of dust emissions (6 bin distributions) in the model. However, sea salt aerosol emissions are not optimized by AOD assimilation, because sea salt aerosols are difficult to distinguish from dust and carbonaceous aerosols, particularly over coastal areas where AOD does not correlate with sea salt (see Fig. S1). For the direct concentration modification, AOD assimilation optimizes total aerosol mass concentrations including sulfate, nitrate, ammonium, carbonaceous, soil dust, and sea salt aerosols, while their aerosol composition and size distribution for soil dust and sea salt (6 and 4 bin distributions, respectively) remains the a priori information obtained from the ensemble forecasts during the analysis step.”

Since the principal novelty of this work is the synergistic use of gas and aerosol observations through ensemble-derived cross-species covariances, the manuscript would benefit from a more explicit description of these update pathways.

Unfortunately, we could not utilize cross-species covariances between secondary aerosol species and their precursor gases to update them, because we do not find strong correlation coefficients between them at 32 ensemble sizes even in polluted areas. Instead, we propagate the observational information on precursor gases into secondary aerosol species through the model response to emission optimization. We clarify these points as follows: (p. 6, l. 170–174)

“We set to zero for error correlations between aerosols and trace gases, such as secondary aerosols and their precursor gases, because the modest correlations between them were found even over polluted areas (Fig. S1). After the emission optimizations, during the next forecast step, sulfate, nitrate, and ammonium, dust and carbonaceous aerosol mass concentrations are indirectly influenced by the updated NO_x, SO₂, dust, carbonaceous aerosol emissions.”

I think cross-species/variables covariance/correlation plots (as in previous Miyazaki et al. papers), or schematic or table explicitly explaining the localization setup would substantially improve the transparency and reproducibility of the methodology. In particular, it would be helpful to indicate which observation types are allowed to update which state variables whether these updates occur directly during the analysis or indirectly during the forecast,

I agree with your suggestion. As done in previous reply, we added the schematic matrix in Fig. 1 and the description that clarify which observed variables are allowed to update which unobserved variables. (p. 6, l. 158–170)

and the physical rationale for retaining some data assimilation couplings while suppressing others.

We also added the reason why we retain carbonaceous aerosol and dust emission optimizations from AOD and suppress sea salt emission optimization from AOD. (p. 6, l. 163–167)

“We supposed primary carbonaceous aerosols and soil dust emissions to be remaining error sources, because the trace gas data assimilation and direct modification of aerosol concentrations through the AOD assimilation reduce model errors unrelated to primary

aerosol emissions. However, sea salt aerosol emissions are not optimized by AOD assimilation, because sea salt aerosols are difficult to distinguish from dust and carbonaceous aerosols, particularly over coastal areas where AOD does not correlate with sea salt (see Fig. S1). ”

Incomplete description of heterogeneous chemistry in methodology

The description of the heterogeneous chemistry requires further clarification. Section 2.1.1 documents only the heterogeneous uptake of HNO_3 on mineral dust, whereas Section 3.3 attributes changes in OH and NO_2 to heterogeneous HO_2 uptake on aerosols. It is therefore unclear which heterogeneous gas–aerosol interactions are represented in the forecast model. This information is important not only for reproducibility but also for interpreting the data assimilation methodology, since the forecast chemistry determines the ensemble-derived cross-species background-error covariances that transfer observational information between gases and aerosols. A more complete description of the heterogeneous chemistry, including the represented gas–aerosol coupling pathways and their role in secondary aerosol formation, would substantially improve the transparency of both the model and the data assimilation framework.

Thank you for pointing out this important thing. The forecast model considers 9 heterogeneous reactions, such as hydrolysis of N_2O_5 and uptake of HO_2 . The direct and indirect modification of aerosol concentrations influence the trace gas analysis during not the analysis step but the forecast steps through these heterogeneous reactions, because we blocked the error correlations between AOD and gas species related to heterogeneous reactions due to limited ensemble sizes. Therefore, we have added Table that summarize heterogeneous reactions on tropospheric aerosols to the Supplement and the following description to the main text:

(p. 3, l. 71–74)

“The model includes 9 heterogeneous chemical reactions on aerosols (Table S1), including a reaction of HNO_3 on the mineral dust surface using ...”

Insufficient discussion on data assimilation constraint versus model response in results

This comment is directly related to the above comment on cross species covariance localization. The manuscript demonstrates that assimilating trace-gas observations improves aerosol composition. However, the discussion does not clearly distinguish between information introduced directly by the observations data assimilation and information propagated indirectly through the model during the forecast step. While the chemical pathways linking precursor gases to secondary aerosols are described, it would strengthen the paper to discuss the information content of the different observing systems. If any, which aerosol species receive independent observational constraints from individual trace-gas measurements, and which improvements arise primarily through the data assimilation covariance versus through the model response? Such a discussion would help readers better understand the origin of the reported synergistic effects.

We appreciate your comments. We insufficiently clarified the aerosol update pathways. First, as described in the reply to the above comment, we do not account for cross error correlations between individual aerosol species and related trace gases because of limited ensemble sizes and weak correlations (Fig. S1). Therefore, aerosol updates through trace gas data assimilation resulted from the model responses to the optimizations of precursor gases. The updates through aerosol data assimilation are subject to the modification of concentrations and emissions (and subsequent model responses). To clarify the respective impacts of direct concentration modification and model response to the optimized emissions, we performed the DA calculation without emission optimizations. The impacts of the model responses play critical roles in aerosol composition estimates, because the direct concentration modification assumes retaining *a priori* information on aerosol compositions in the present system. We have added the corresponding description as follows:

(p. 12, l. 373–p. 13, l. 378)

“To quantify the respective contributions of direct concentration modification and emission optimization to the aerosol analyses, we conducted the sensitivity DA calculation without emission optimization (Table 2 and Fig. 2). The modifications of all the aerosol components in the Emis-fixed DA were reductions for most cases. The modifications in the GasAero DA varied between different aerosol components. The modifications of nitrate and carbonaceous aerosol analyses in the GasAero DA showed the opposite signs against those in Emis-fixed DA. These results demonstrated that the emission optimizations play critical roles in the aerosol composition estimates.”

Somewhat limited discussion of regional validation

The manuscript focuses primarily on cases where the coupled assimilation improves the analysis, but several validation metrics show degraded performance, particularly for European nitrate aerosols and some correlation coefficients. While some explanations are provided (e.g., limited AOD coverage or uncertainties in transport and deposition), these cases deserve a more detailed discussion because they may reveal important limitations of the coupled gas–aerosol assimilation framework. In particular, it would be useful to explain why GasAero performs worse than Aero-only for European nitrate and why some correlation coefficients decrease despite reductions in bias or RMSE.

According to comments from anonymous reviewer #1, we have added the validation of aerosol precursor gases. The validation shows that GasAero DA reduced negative biases in surface NO_2 , even though positive biases in nitrate aerosols were increased by data assimilation through the model response. These results suggest that nitrate aerosol concentrations cannot be improved not only by NO_x emission optimization. These positive biases can partially be attributed to N_2O_5 hydrolysis with the default value of N_2O_5 uptake coefficient (0.1), which is much higher than the recommended value from recent studies (0.002–0.02) (Chang et al., 2016; Phillips et al., 2016; Wang et al., 2020). We have added the following discussion in Section 3.

(p. 16, l. 501–504)

“As an exception, the performance on nitrate aerosols became worse through trace gas data assimilation over Europe, even though reducing NO_2 model biases. These positive biases can partially be attributed to nitrate formation through N_2O_5 hydrolysis with the default value of uptake coefficient of 0.1, which is much higher than recent recommended values of 0.002–0.02 (Chang et al., 2016; Phillips et al., 2016; Wang et al., 2020).”

Understanding these cases is important for identifying the situations in which the coupled assimilation is beneficial and those in which it may introduce undesirable cross-species corrections.

As previous replies, positive biases in nitrate aerosols were degraded when we optimized precursor emissions rather than direct concentration modification. Even when we block the cross-error correlation between NO_x and nitrate aerosols, we could not completely remove the degradation effect of model biases in related processes. Because cross error correlations between different aerosol and gas species are generated by ensemble model forecasts, we

expect further degradation of nitrate analysis when we consider the error correlation between NO_x and nitrate aerosols. Either way, the model performance on aerosol-gas interaction processes is essential for improving the aerosol-gas coupled data assimilation.

We mention this point in Section 6.2 as follows:

(p. 21, 1, 657–660)

*“Model misrepresentation of aerosol-gas interactions through heterogeneous chemistry could degrade aerosol analyses and cross-variable error correlations. Such model developments will be necessary to enhance the data assimilation performance **utilizing cross-variable error covariances** in future studies.”*