

Reply to Comments by Anonymous Referee #1 on:

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

Takashi Sekiya, Kazuyuki Miyazaki, Henk Eskes, Pieter Rijsdijk, Kengo Sudo, and Yugo Kanaya

EGUsphere [preprint], <https://doi.org/10.5194/egusphere-2026-1681>

Received: 25 Mar 2026 – Discussion started: 27 Apr 2026

We would like to thank anonymous referee #1 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.

The study presents a comprehensive comparison of different satellite retrieval assimilation strategies utilising the CHASER V4.0 Chemical Transport Model (CTM) with a Kalman Filter approach. This data assimilation (DA) method is specifically designed to simultaneously adjust both atmospheric concentrations and emissions, thereby enabling a more accurate representation of atmospheric composition.

The analysis focuses on two principal groups of assimilated observations: gas-phase retrievals and aerosol optical depth (AOD) measurements. The gas-phase retrievals include tropospheric column measurements of nitrogen dioxide (NO₂), total column sulphur dioxide (SO₂) from the Ozone Monitoring Instrument (OMI), carbon monoxide (CO) from MOPITT, and stratospheric profiles for ozone (O₃) and nitric acid (HNO₃) from the Microwave Limb Sounder (MLS). In addition, AOD measurements are incorporated using data from MODIS.

The findings indicate that the combined assimilation of trace-gas and AOD observations yields the most robust results. This outcome is validated through evaluations using AERONET AOD readings, surface particulate matter (PM) data from Europe, North America, and China, and results from the KROKOS aircraft campaign. Furthermore, the authors explore the impact of assimilating data from TROPOMI and VIIRS in place of OMI and MODIS, providing additional insight into the effectiveness of different satellite retrieval combinations.

We appreciate your careful review and positive comments.

General comments:

The paper demonstrates the efficacy of a state-of-the-art data assimilation (DA) system for multiple satellite retrievals of atmospheric composition, capable of constraining both emission fluxes and atmospheric concentrations. The system exhibits robustness, as incorporating additional and complementary satellite observations enhances the realism of concentration field analyses and subsequent forecasts.

Thank you for positive comments.

The authors observe that their DA system tends to adjust emissions more than concentrations, warranting further discussion. Specifically, since the operation of any DA system is influenced by the selection of prior uncertainties or perturbations during propagation within the Kalman filter framework, it would be beneficial to explore how these parameter choices affect the relative adjustments to emissions and concentrations.

We additionally conducted sensitivity assimilation calculations with 50% and 100% emission perturbation reductions relative to standard settings (Figs. S8 and S9). In the calculation with 50% emission perturbations, the modifications of emission and concentration analyses were substantially decreased. The DA run with 100% emission perturbation reductions show much smaller modification in aerosol concentrations than those in the DA run without emission optimization. These results demonstrate that emission perturbations play essential roles in both the data assimilation analysis of aerosol emissions and concentrations. The following discussion has been added to the main text:

(p. 18, l. 538–545)

“Selection of predefined minimum emission spreads for inflation was essential to modify the emissions and concentrations of aerosols and its precursors (Figs. S8 and Table S6). By

reducing the initial and predefined minimum emission spreads by 50% and 100%, emission modifications made to data assimilation were decreased by up to 0–53% and 100%, respectively, for carbonaceous, dust, NO_x, and SO₂ emissions. The mean PM_{2.5} concentration modifications over eastern China with 50% emission perturbation reductions were decreased by 13%. The concentration modifications with 100% emission perturbation reductions (by 15%) were much smaller than those in the Emis-fixed DA (by -63%). These results suggest that the selection of emission perturbation parameters plays critical roles in not only emission optimization and subsequent model responses but also direct concentration modifications.”

A tabulated summary detailing the assumed or typical calculated uncertainties for emission fluxes and mass mixing ratios would provide valuable clarity.

We added a table that summarizes uncertainty in emissions and concentrations to Supplement (Table S5).

Reporting on posterior emissions should be contextualized alongside prior uncertainties.

We have added the statement on the emission analysis spread as follows:

(p. 17, l. 515–518)

“The regional mean analysis spreads of carbonaceous emissions over China and Southeast Asia were reduced to approximately 30% compared to the assumed initial uncertainty (67%). As an exception, the analysis spread over India exceeded 100%, indicating the lack of observational constraints associated with these sparse spatial coverages of AOD retrievals in monsoon season.”

(p. 17, l. 523–525)

“The mean analysis spreads of dust emissions were relatively high compared to those in carbonaceous aerosols over most regions, because the spatial coverage of AOD with α of >0.5 used for constraining dust emissions was small.”

(p. 17, l. 529–530)

“The regional mean analysis spreads of SO₂ emissions ranged from 3 to 17%, which was smaller than the assumed initial uncertainty (32%).”

(p. 17, l. 535–537)

“The regional mean analysis spreads of NO_x emissions over most regions were reduced

from the assumed initial uncertainty (56%) to below 45%, except for Central Africa and South America.”

Additionally, because both concentrations and emissions are subject to modification, updates to emissions alone cannot offer a comprehensive commentary on emission fluxes; issues related to emissions may still manifest as concentration changes.

Thank you for pointing it out. Both AOD DA directly corrects total aerosol concentrations and trace gas DA indirectly corrects secondary aerosol components. Conceptually, we suppose that these corrections lead to improved primary aerosol emission optimization through reducing model errors unrelated to primary aerosol emissions. We clarified this point as follows:

(p. 4, l. 103–106)

“This extended system takes individual advantages of AOD and trace gas DA. Specifically, it improves primary aerosol emission optimization, because AOD DA reduces model errors unrelated to emissions through direct aerosol concentration modification and trace gas DA reduces model errors related to secondary aerosol components through precursor gas emission optimizations.”

Based on the results of this study, primary aerosol emission updates, especially for carbonaceous emissions, were influenced by secondary aerosol concentrations, whereas dust emissions were driven by AOD assimilation. We clarified these points in the main text as follows:

(p. 17, l. 513–515)

“The slight differences between the GasAero and Aero-only DA runs were due to small AOD reductions in most regions resulting from reduced sulfate aerosol concentrations due to SO₂ emission reductions.”

(p. 17, l. 522–523)

“These corrections made by the GasAero DA were largely driven by the Aero-only DA, because dust aerosol is a dominant component of total aerosols over major desert regions.”

A methodological limitation—either inherent to the method or its description—appears to be the influence of Aerosol Optical Depth (AOD) assimilation on primary and secondary aerosol component concentrations. While all components affect AOD, only the surface fluxes of primary dust ($AE > 0.5$) and carbonaceous aerosol ($AE < 0.5$) seem modifiable.

Sea salt aerosol emissions are not optimized by AOD assimilation, because it is difficult to distinguish dust and carbonaceous aerosols from sea salt aerosols especially over coastal areas where AOD does not correlate with sea salt. The above statement was added to the main text (p. 6, l. 165–167).

Clarification on the distribution among the three dust bins, and the consideration of sea salt bin fluxes, is necessary.

We retain the *a priori* distribution among 6 dust bins and 4 sea salt bins for concentrations (both dust and sea salt) and emissions (only for dust). We described these points in the main text as follows:

(p. 6, l. 162)

“... while we remain the assumption of size distribution of dust emissions (6 bin distributions) in the model.”

(p. 6, l. 168–170)

“... 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.”

It remains uncertain why the AOD assimilation is designed to only correct dust and carbonaceous aerosol.

We clarify the reason why we select carbonaceous and dust aerosol emissions for emission optimization through AOD assimilation as follows:

(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)."

Greater detail regarding the specific impacts of AOD assimilation on individual aerosol concentrations and their respective fluxes is required.

We updated the description how AOD assimilation corrects individual aerosol concentrations and fluxes. For individual concentration corrections from AOD assimilation, we retain a priori aerosol composition derived from the ensemble forecast. All the aerosol components are perturbed for AOD assimilation.

(p. 6, l. 167–174)

"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 the 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. We set to zero for error correlations between aerosols and trace gases, such as secondary aerosols and their precursor gases, because strong correlations between them were not 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 , SO_2 , dust, carbonaceous aerosol emissions."

Carbonaceous aerosol and soil dust emissions are subject to flux corrections from AOD assimilation as we explain in the above reply.

(p. 6, l. 158–162)

"Specifically, as summarized in Fig. 1, we used ... 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."

To show the combined and respective impacts of AOD and trace gas data assimilation on individual aerosol components, we have added impacts on carbonaceous, dust, and sea salt aerosols to Figure 1 and the corresponding descriptions (p. 12, l. 357–361).

Evaluation using in-situ data is incomplete. Since NO_2 , SO_2 , and O_3 measurements are assimilated, the analyses should also encompass surface air quality (NO_2 , SO_2 , and O_3)

observations from European, North American, and Chinese networks, similar to the approach used for PM_{2.5}.

We added the validation of NO₂, SO₂, and ozone using air quality observation networks, such as AQS, EEA AQ e-reporting, and Japanese network and AirKorea to Table 3 and Figures S4–6, which is same as PM_{2.5} validation. The corresponding descriptions were also added to the main text:

(p. 15, l. 445–449)

“As shown in Fig. S4, CTL overestimated the observed SO₂ over eastern United States and underestimated NO₂ over the source areas. GasAero DA reduced the positive SO₂ and negative NO₂ mean model biases compared to the AQS network. For NO₂, negative model biases over the eastern United States became small positive biases after GasAero DA, while positive model biases were reduced over the western United States. Surface ozone concentrations in CTL were overestimated. GasAero DA degraded the positive model biases across the United States. These changes were dominated by Gas-only DA.”

(p. 16, l. 468–473)

“CTL overestimated the observed SO₂ over the eastern Europe, except for Romania and Bulgaria (Fig. S5). GasAero DA reduced the positive model biases, whereas slightly degraded the negative model biases over Romania and Bulgaria. The surface NO₂ concentrations in CTL were underestimated across Europe. These negative model biases were improved by GasAero DA for most cases, while small negative biases over United Kingdom, Belgium, and Netherlands became positive biases. Positive model biases of surface ozone across Europe were found in CTL. GasAero and Gas-only DA similarly degraded the positive biases.”

(p. 16, l. 489–495)

“CTL overestimated the observed SO₂ over the Seoul Metropolitan and Greater Tokyo areas (Fig. S6). GasAero DA substantially reduced these biases. NO₂ model biases over South Korea were positive, whereas those over Japan were negative. GasAero DA reduced positive model NO₂ biases over South Korea, whereas positive model biases were increased by Gas-only DA. Over Japan, negative model biases remained in GasAero DA, while Gas-only reduced negative model biases. Meanwhile minor changes in NO₂ biases were found in Aero-only DA. These results implicate that aerosol–trace gas interaction, such as N₂O₅ hydrolysis on aerosols, reduces NO₂ concentrations. Surface ozone concentrations in CTL were overestimated. GasAero DA degraded positive biases.”

The evaluation appears to focus primarily on analysis of the concentration. Including an assessment of multi-day forecasts with optimized emissions could further motivate the significance of emission inversion.

We appreciate this helpful suggestion. Although the developed system could improve short-term predictions through optimized emissions, our data assimilation system has focused on developing atmospheric chemistry reanalysis data (e.g., Miyazaki et al., 2020), rather than improving air quality forecast. The component described in this manuscript is going to be included in the new version of atmospheric chemistry reanalysis. While assessing air quality predictability is beyond the scope of the present study, the following statement has been added to discuss potential application:

(p. 22, l. 704–p. 23, l. 707)

“... A lack of information on PM_{2.5} compositions and emissions could also limit the performance of air quality forecasting. Long-term reanalysis datasets based on the simultaneous assimilation of aerosol and trace-gas observations can provide observationally constrained aerosol speciation information to address these limitations.”

In the evaluation section, performance statistics are frequently presented both narratively and in tables, resulting in unnecessary duplication. Retaining the tables while consolidating textual references is advisable.

The duplications were removed and the main texts were condensed in Sections 3.2, 3.3, and 3.4.

The comparison between OMI/MODIS and TropOMI/VIIRS could be moved to supplementary materials, as it does not fundamentally enhance the primary discussion on combined AOD and gas retrieval DA and inversions.

Thank you for this suggestion. Since our data assimilation system is designed to produce atmospheric chemistry reanalysis, it is essential to characterize how changes in observing system affect data assimilation performance. Therefore, we have decided to retain this comparison in the main texts, while clarifying its motivation and finding as follows:

(p. 19, l. 583–584)

“It is essential to characterize impacts of observing system changes on data assimilation performance for developing long-term chemical reanalysis.”

(p. 20, l. 621–623)

“Overall, the results suggest benefits of assimilating current generation satellite constellations for constraining sulfate aerosols and AOD. For nitrate, the current constellation improved agreements with EANET compared to the previous constellation, while the current constellation reduced RMSEs for PM_{2.5} over the United States and Europe.”

(p. 21, l. 643–645)

“We also characterize the impacts of assimilating the current-generation satellite constellations (i.e., TROPOMI and VIIRS) compared to the earlier-generation constellation (i.e., OMI and MODIS), since assessment of observing system changes is critical for developing long-term reanalysis data.”

Results indicate both improvements and degradations with the newer observations, which should be clearly communicated.

We clarified the description on the degradation of DA performance due to assimilating the current satellite constellation compared to the previous constellation:

(p. 19, l. 600–602)

“These bias reductions were larger than those from OMI+MODIS DA. RMSEs for TROPOMI+VIIRS DA were comparable to or 16–29% smaller than those from OMI+MODIS DA for ATom-4, CASTNET, and EMEP, **although the RMSE relative to EANET was 2% larger.**”

(p. 19, l. 606–607)

“These bias and RMSE reductions were smaller than those from OMI+MODIS DA for EMEP and CASTNET but larger for EANET.”

(p. 19, l. 608–609)

“RMSEs in TROPOMI+VIIRS DA ..., but 29% larger for ATom-4.”

(p. 19, l. 612–613)

“The bias and RMSEs in TROPOMI+VIIRS DA were also degraded, compared to those in OMI+MODIS DA.”

(p. 19, l. 618–620)

*“The bias reductions in TROPOMI+VIIRS DA were comparable to or **smaller than those from OMI+MODIS DA**. RMSEs for TROPOMI+VIIRS DA were 0.3–8% smaller than those in OMI+MODIS DA for AQS and EEA, **but 11% larger for the Japanese and AirKorea networks.**”*

Chapter 6 (Discussion and Summary) needs to be streamlined. Conclusions—including findings and future directions—are very brief, and topics such as insufficient constraints on vertical profiles could be addressed.

Thank you for pointing it out. We mentioned the vertical aerosol profiles as an example of insufficient constraints in the present DA system:

(p. 21, l. 663–666)

“As shown in the comparison with aircraft observations, the present DA system insufficiently constrain the vertical structures, because AOD observations explicitly have no information on vertical aerosol profiles. Therefore, vertical aerosol extinction profiles from CALIOP (Winker et al., 2013) and EarthCARE/ATLID (van Zadelhoff et al., 2023) offer valuable information on aerosol layer heights.”

The ensemble’s limited capacity as such to provide sufficiently large emission uncertainty is another issue worth discussing.

We also discuss the limited capacity of the present DA system to estimate background uncertainties:

(p. 22, l. 688–696)

*“Incorporating cross-variable background error covariances between an expanded set of state variables, including aerosol optical properties (AOD, SSA, AE, and **vertical profiles of extinction coefficients**) and individual aerosol components, would improve the DA system’s ability to exploit observational information by propagating it across a broader state vector **across horizontal, vertical, and inter-variable directions**. Nevertheless, achieving these improvements requires accurate error covariance estimates. **The present DA system has limited capacity to estimate accurate background error covariances. Most emission inventories lack information on uncertainties, and uncertainties in the forecast models’ transport, chemistry, and deposition processes prevent the accurate propagation of emission errors. In most cases, inherent model process errors have not been***

quantified. The limited ensemble size also leads to the sampling errors of background error covariances. Important technical challenges remain as mentioned above.”

On the other hand, there is no need for an extensive overview of alternative or forthcoming satellite products (Section 6.3) in this paper; new satellites should only be mentioned if they address concerns directly raised in the manuscript.

We omitted some of mentions on specific satellites products and condensed Section 6.3. However, discussions on development of future observing networks for aerosols based on the conclusions of this study play a central role in the present study. Thus, we have kept many parts of Section 6.3 in the revised manuscript (p. 21, l. 670–p. 22, l. 671).

Similarly, the model performance section (6.2) can be condensed.

We also condensed this part to one paragraph as follows:

(p. 21, l. 651–660)

“The performance of data assimilation is influenced by uncertainties in the forecast model, particularly associated with aerosols and their precursors. For example, the relationship between aerosol mass concentrations and AOD depends on aerosol optical parameters (e.g., mass extinction coefficient and single scattering albedo), which are strongly influenced by aerosol microphysical properties. Furthermore, uncertainties in the model deposition and chemical reaction processes of aerosol precursors, such as NH_3 , HNO_3 , and SO_2 , can influence the estimates of precursor emissions, aerosol formation, and aerosol composition. Although the forecast model does not include explicit representations of the NH_3 volatilization processes over land, implementing these processes could improve the model performance for simulating NH_3 (Zhu et al., 2015; Cao et al., 2022). Model misrepresentation of aerosol-gas interactions through heterogeneous chemistry could degrade aerosol analyses even when we suppress cross-variable error correlations. Such model developments will be necessary to enhance the data assimilation performance utilizing cross-variable error covariances in future studies.”

The brief “Climate and Health” section (6.4) may be omitted, as it contributes little new information specific to this paper.

We merged this section with conclusion section to state the future potential application of the present DA system as:

(p. 22, l. 698–p. 23, l. 707)

“Overall, our results demonstrate that the simultaneous assimilation of satellite AOD and trace gas observations provides clear advantages ... **Because different aerosol components have distinct climatic effects (Li et al., 2022a), their evaluation is important, yet till limited by sparse observational coverage. Likewise, mortality risks associated with PM_{2.5} exposure can be estimated more accurately when chemical composition is considered, rather than using only total PM_{2.5} mass as in many previous studies (Beelen et al., 2015; Ueda et al., 2016; Thurston et al., 2016; Weichenthal et al., 2024; Crouse et al., 2016; Xue et al., 2021). A lack of information on PM_{2.5} compositions and emissions could also limit the performance of air quality forecasting. Long-term reanalysis datasets based on the simultaneous assimilation of aerosol and trace-gas observations can provide observationally constrained aerosol speciation information to address these limitations. Future improvements ...**”

Specific comments:

L8 clarify “conventional”

Conventional DA runs are defined on L6 as “conventional DA runs that separately assimilate AOD or trace gases satellite observations.”

L51 The CAMS re-analysis (Inness et al. 2019, Flemming et al. 2017) assimilate trace gases and AOD simultaneously, although not including the update of emissions.

We have added the comment on the CAMS reanalysis as
(p. 2, L. 42–43).

“The CAMS (EAC4) reanalysis data simultaneously assimilates AOD and trace gas observations, although it does not estimate aerosols and their precursors’ emissions (Inness et al., 2019; Garrigues et al., 2022).”

L74 Sea salt is part of the model but seems to be not included in the DA framework. Why ?

Sea salt aerosol concentrations are subject to modify due to AOD data assimilation, whereas sea salt emissions are not optimized. See the reply to general comments for the details.

L 107 Please clarify if x constrains both the concentrations and the emissions. Please provide more information about your framework to optimise emissions

We have added the descriptions on x in this paragraph as

(p. 5, l. 120–121)

“State vector x includes 35 gaseous chemical species, 7 aerosol species, surface NO_x, CO, SO₂, carbonaceous aerosols (i.e., sum of BC+OM), and soil dust emissions.”

L 117 Please explain the self-consistency test here

We have condensed and moved the result of self-consistency check in the Supplement to the main text (p. 5, l. 124–128).

L 127 Does this mean the ensemble systematically underestimated the emissions uncertainty? What is the motivation of the minimum values and where do the number come from?

Yes, this suggests a systematic underestimation of the emission uncertainty. Because no forecast model applied to the emissions, the ensemble spread without covariance inflation tends to underestimate the time evolution of emission uncertainty. We implement covariance inflation to the predefined minimum values to prevent these underestimations of emission uncertainty. We set the predefined values for carbonaceous aerosol and dust emissions according to the sensitivity calculations as summarized in Table S1. The predefined values for NO_x, CO, and SO₂ emissions were taken from our previous studies (e.g., Miyazaki et al., 2020). We have revised this part to clarify emission inflation as follows (boldface indicated the added parts):

(p. 5, l. 134–139)

*“In the analysis step, the standard deviation of the emission ensembles was ... to prevent covariance underestimation **because of the absence of any forecast model to the emissions (Miyazaki et al., 2012).** ... The minimum values for **primary** aerosol emissions were determined from the sensitivity calculations (Table S1), **while those for trace gases were taken from our previous studies (e.g., Miyazaki et al., 2020a).**”*

L 131 Why is sea salt no considered here ?

This is because it is difficult to distinguish dust and carbonaceous aerosols from sea salt aerosols. See the reply to general comments for the details.

L 153 Please check for duplication of the information in section 2.2 and table 1. Having the info in the table should be sufficient

We removed the information duplicated to Table 1.

L 263 Table 2 also contain the info about MLS.

We added the information on MLS to Table 2.

L 263 Please modify Fig 1 to clarify that only sea salt and OC are optimized.

The present DA system directly corrected individual aerosol concentration through AOD assimilation, while keeping a priori aerosol composition derived from the ensemble forecast. Meanwhile, we designed the present system to optimize only carbonaceous aerosol and soil dust emissions by AOD assimilation, because the trace gas data assimilation and direct modification of aerosol concentrations through the AOD assimilation reduce model errors unrelated to primary aerosol emissions and because fraction of sea salt aerosols in AOD are difficult to distinguish from dust and carbonaceous aerosols, particularly over coastal areas (see also the reply to general comment). To clarify this point, we modified Fig. 1 as follows: (1) we divided primary aerosol emissions into three categories, such as carbonaceous, dust, and sea salt; (2) we applied sky blue shading to the categories that is subject to the emission optimization in the present system.

L 285 Please compare also against the surface NO₂, SO₂ and O₃ observations from the AQ networks (as for PM_{2.5})

We added validation results for NO₂, SO₂, and O₃ with AQ observation networks which is same as PM_{2.5}. (See the reply to general comment for more details.)

L 301 Please specify the location of the KORUS campaign

We added the location of KORUS-AQ campaign:

(p. 11, l. 327)

“The KORUS-AQ flights covered 124–133°E and 30–38°N (Fig. 5)”.

L 324 It remains unclear how you attribute the concentration analysis changes to emissions modification and direct concentration updates (see general comment)

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 remains the a priori information during the analysis step. The present DA system is designed to optimized emissions of primary carbonaceous aerosols and soil dust, 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 relative fraction of sea salt aerosols in AOD are difficult to distinguish from dust and carbonaceous aerosols. 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.

As described in the reply to the general comment, we updated the corresponding description in Section 2.2 (p. 6, l. 167–174)

L333 Why is this the case. What choices have been made to suppress the impact.

We don't assimilate NH₃ observations to optimize NH₃ emissions in the present system because the model has relatively large forecast errors in NH₃, in part because it does not explicitly represent NH₃ volatilization. These errors could introduce large biases into the SNA aerosol analyses. To assess this potential impact, we adjust NH₃ emissions based on the difference between model-simulated NH₃ and CrIS satellite observations. We clarified these part as follows:

(p. 12, l. 352–356)

“Furthermore, we assessed the potential impact of NH₃ emission biases on SNA aerosol analyses. NH₃ observations are not assimilated to optimize emissions in the present system, because forecast model errors are relatively large partly owing to the lack of an explicit representation of NH₃ volatilization. We adjusted NH₃ emissions based on the difference between model-simulated NH₃ and CrIS observations. This adjustment increased nitrate and ammonium aerosols (Fig. S3), with little effect on sulfate (<2%), consistent with thermodynamic equilibrium among SO₄²⁻, NO₃⁻, and NH₄⁺.”

L 341 Please clarify how this finding was derived. The attribution seems a bit ad hoc.

The attribution is based on the comparison of the Gas-only and Aero-only DA runs. We have clarified these part as follows:

(p. 12, l. 368–371)

“The Gas-only and Aero-only DA runs suggest that corrections to sulfate, nitrate, and ammonium from assimilating trace gas observations were larger than those from AOD assimilation over most regions. As an exception, impacts of AOD assimilation on sulfate and ammonium were larger over North and South Africa, suggesting that uncertainties in aerosol formation, transport, and deposition processes (e.g., heterogeneous reactions on primary aerosols) are larger than those in precursor gas emissions.”

L 351. Please clarify this explanation.

We have added the reason why the control simulation shows lower mean biases and DA does not tend to improve the negative model biases at sites with high aerosol concentrations as:

(p. 13, l. 380–389)

“..., biases in the control model simulation at individual sites largely varied between -0.27 to 0.76, and canceled out each other, resulting in relatively small mean bias. ... AOD DA reduced positive model biases and small negative biases in most cases, which improve correlation coefficients and RMSEs. However, large negative model biases at sites with high aerosol concentrations, such as East Asia, South Asia, the Middle East, and the Saharan Desert, tend to be degraded through AOD DA, likely because enhancement of the observed AOD cannot be represented in the coarse-resolution model.”

Say more clearly that Aero DA improved R and RMSE but slightly degraded the biases.

As stated in the above reply, the control simulation shows lower mean biases apparently, resulting from compensation of positive and negative model biases at individual sites. AOD DA reduced these biases, except for sites with large negative biases. The tendency resulted in improved R and RMSE due to AOD DA with slight degradation of mean biases.

L 356 Please discuss in this section the inability of the DA system to modify the profile shape – or the opposite.

We have added a discussion on vertical profile shapes:

(p. 13, l. 407–p. 14, l. 410)

“Overall, improvements in aerosol concentrations were evident in the lower troposphere. During forecast steps, top-down estimates of aerosols and their precursor emissions influenced aerosol concentrations primarily in the lower troposphere, whereas direct AOD assimilation adjusted concentrations without changing the shape of their vertical profiles.”

L 458 Please clarify how this was identified? .

To identify this more quantitatively, we added the sensitivity DA calculation without emission optimization to Fig 2. We removed this sentence here and add a paragraph to explain the impacts of direct concentration modification and emission optimization on concentration analyses 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.”

L 485 Please discuss here more clearly that your experiments only cover the summer period.

We have added a discussion on the period selection:

(p. 18, l. 552–554)

“The comparison was conducted for May-August 2016 when precursor emissions affect secondary aerosols under photochemically active environment over the major polluted regions.”

Has this taken into account in the comparison of the emissions totals?

Yes. We mentioned this in the revised manuscript:

(p. 18, l. 554)

“The recent emission inventory was also averaged during the same periods.”

L 570 The summary should also contain the OMI/MODIS vs TropOMI/VIIRS comparison

We added the paragraph on the comparison between OMI/MODIS and TROPOMI/VIIRS DA to Section 6.1:

(p. X, l. X)

“We also characterize the impacts of assimilating the current-generation satellite constellations (i.e., TROPOMI and VIIRS) compared to the earlier-generation constellation (i.e., OMI and MODIS), since assessment of observing system changes is critical for developing long-term reanalysis data. The use of TROPOMI and VIIRS observations benefited for constraining sulfate aerosols and AOD due to assimilating the current constellation compared to the previous constellation. Improvements were found for nitrate against EANET and for PM_{2.5} against air quality networks over Europe and the United States. The current constellation can produce long-term records in aerosol composition reanalyses together with earlier-generation datasets such as OMI and MODIS.”

For section 5 and 6 – please see my general comment. They should be completely reviewed.

Section 5 has been condensed, but remained in the main text, because the assessment of observing system changes is essential to develop long-term reanalysis dataset, as discussed in the revised manuscript, as above reply to general comments.

According to above general and specific comments, we have condensed and revised section 6.

Figure 5: Please add time and location of the profile observations

We added a map of KORUS-AQ campaign flight paths on individual days with different colors to Figure 5.

Figure 6: Why are the PM 2.5 no circles but grid points ?

The spatial coverage of the observation networks for PM_{2.5} as well as SO₂, NO₂, and ozone were denser than those for sulfate, nitrate, and ammonium. Therefore, for PM_{2.5}, it is difficult to see the model biases at individual sites, because multiple observations exist at each model grid point. We compare model and observations at model grid points. We have added the following sentence:

(p. 10, l. 305–306)

“The observation networks for PM_{2.5} as well as SO₂, NO₂, and ozone were denser than those for sulfate, nitrate, and ammonium. We compare model and observations at model grid points.”

Table 2: Mention the MLS observations

We included MLS assimilation or not in Table 2.