Authors’ Response to Reviews of

The sensitivity of aerosol data assimilation to vertical profiles: case study of dust storm assimilation with LOTOS-EUROS v2.2

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RC: ***Reviewers’ Comment***, AR: Authors’ Response, ☐ Manuscript Text

1. Overview

RC: ***The manuscript "The sensitivity of aerosol data assimilation to vertical profiles..." by Pang et al. investigates the sensitivity of assimilated vertical aerosol profile accuracy to the prior model state generated by atmospheric models. The authors perform data assimilation using the PyFitter toolbox, a stochastic Ensemble Kalman Filter (EnKF) implementation developed by the authors, using ground-based PM_{10} and Himawari-8 satellite-derived dust optical depth (DOD) data. Through several case studies of dust storms, it shows that while assimilation of accurate profiles improves the consistency of 3D aerosol states, flawed priors can exacerbate vertical structural errors. Validation using CALIPSO and LiDAR confirms these findings, emphasizing the need for precise vertical profiling.***

While the manuscript is well organized, with a clearly written methodology supplemented by appropriately captioned plots, I think there are several major areas where both the science and writing need to be revised before publication in GMD. Once these are addressed, I would be happy to review the manuscript and re-evaluate its suitability.

AR: We would like to extend our sincere gratitude to the reviewer for the meticulous and insightful comments provided regarding our manuscript. We assure the reviewer that we have carefully considered each point raised and have made diligent efforts to address them comprehensively within the revised version of our manuscript.

2. Major comments

RC: ***– Please justify how the model state assimilated with DOD data in the optical wavelength (550 nm) can be compared with extinction data collected by LiDAR at a slightly lower wavelength (532 nm)***

AR: Thanks for the comment. The relationship between dust extinction coefficient and DOD is not explained in the original manuscript. We have added a paragraph about the conversion of AOD under different wavelengths in the Appendix. And a new section about how to calculate DOD from extinction coefficient is added for further clarity. The wavelengths $550\,\mathrm{nm}$ and $532\,\mathrm{nm}$ are very close in the visible spectrum, with a difference of only $18\,\mathrm{nm}$. Dust optical properties, such as extinction coefficient, do not vary significantly over such a small wavelength range. This allows for a reasonable approximation when comparing data at these two wavelengths.

Conversion of AOD between different wavelengths
One common method for converting AOD between different wavelengths involves the Ångström exponent ($\mathring{A}$) (Jin et al., 2023). This exponent describes the relationship between AOD and wavelength, defined as the ratio of the logarithm of the AOD ratio at two different wavelengths to the logarithm of the ratio of those wavelengths:

$$\mathring{A} = -\frac{log(\tau_{\lambda_1}/\tau_{\lambda_2})}{log(\lambda_1/\lambda_2)} \quad (1)$$

where: $\mathring{A}$ is the Ångström Exponent. τ_{λ_1} and τ_{λ_2} are the AOD values at wavelengths λ_1 and λ_2, respectively.

Given the AOD at a specific wavelength and the Ångström Exponent, the AOD at another wavelength can be estimated using the following formula:

$$\tau_\lambda = \tau_{\lambda_0}(\frac{\lambda}{\lambda_0})^{-\mathring{A}} \quad (2)$$

Here, λ_0 represents the reference wavelength where the AOD is known, and λ is the target wavelength. The wavelengths $550\,\mathrm{nm}$ and $532\,\mathrm{nm}$ are very close in the visible spectrum, with a difference of only $18\,\mathrm{nm}$. Dust optical properties, such as extinction coefficient, do not vary significantly over such a small wavelength range. This allows for a reasonable approximation when comparing data at these two wavelengths.

DOD operator

Mie theory is applied to convert the aerosol mass concentration into AOD. It is calculated through the scatter and absorption coefficients of spherical particles with a given radius and refractive index (Gupta et al., 2018). Is defined as:

$$\tau = \sum_{k=1}^{n} \epsilon_d^k z^k \quad (3)$$

where τ is the simulated AOD. ϵ_d^k and z^k are the dust extinction coefficient and layer thickness at the kth layer. ϵ_d^k is calculated by the product of extinction efficiency Q_{ext}, total cross section per unit mass S (m^2g^{-1}) and the aerosol mass concentration C (gm^{-3}):

$$\epsilon_d^k = Q_{ext}SC \quad (4)$$

where Q_{ext} is the sum of scattering and absorption efficiency. It's decided by the ratio of aerosol radius, incident wavelength and chemical composition (H. C., 1958). S depends on the particle size and aerosol mass density. The dust bins and diameter ranges are shown in table 1. Detailed descriptions concerning the calculation of AOD can be found in Section 2.2, Jin et al. (2023).

Table 1: Dust size bins and diameter ranges

Bins	dust_ff	dust_f	dust_ccc	dust_cc	dust_c
Diameter range (μm)	0.01-1	1-2.5	2.5-4	4-7	7-10

RC: ***– How is DOD calculated from the Angstrom coefficient and AOD? The authors should move the contents of the supplementary section to the main text since the former is only 1 page and contains useful information***

AR: Thanks for the comment. The descriptions about the calculation of Himawari DOD have been moved into the main text.

To remove the fine-mode non-dust AOD in total AOD, an empirical function concerning Ångström exponent ($\mathring{A}$) is used to calculate the sub-micron fraction (SMF) (Anderson et al., 2005; Di Tomaso et al., 2022). Then the dust optical depth (DOD) can be obtained by the SMF.

$$SMF = -0.0512 \times \mathring{A}^2 + 0.5089 \times \mathring{A} + 0.02 \quad (5)$$

$$DOD = AOD \times (1 - SMF) \quad (6)$$

Furthermore, threshold of $\mathring{A} \leq 1$ is set to exclude the fine-mode dominant observations.

RC: ***– Why is the validation only performed for a single time step in Figs. 3-4, 6-7? The authors should consider providing some measure of total error (average difference between observed and assimilated dust concentration profile) over a pre-defined time window***

AR: Thanks for the comment. The CALIPSO data is sparse in time. It's polar-orbited and only scans through the studied domain for 2 to 3 times a day. The scan time can only be at nighttime to avoid the impact the sunshine. Hence only a single time step is used to validate the assimilation performance. Besides, we have calculated the correlation coefficient (R) across the altitude and latitude as the evaluation metric. To align different vertical resolutions between model and CALIPSO observations, concentration fields from model are liner interpolated into the CALIPSO data point. Metrics on all cases are calculated. Figures and descriptions are added in the Supplementary.

Statistical evaluation
Figures 1 and 2 are the altitudinal and latitudinal trend of correlation coefficient (R) between concentration fields and CALIPSO observations. The fields are linear interpolated into the CALIPSO data point. These data are paired within the altitudinal and latitudinal direction. In the case of *P-Gd-CAL* and *N-Gd-CAL*, as shown in figs. 1 and 2 (a,b), it can be noticed that ground data assimilation can optimize the dust field under the correct vertical structure. Improvements of R can be seen in the 1st case. Meanwhile, as figs. 1 and 2 (b) shows, it can also maintain the incorrect vertical structure. In the case of DOD assimilation, similar trend is shown. The incorrect vertical structure remains after assimilation. Moreover, there is a noticeable degrade of R found in fig. 2 (c), which is in line with the inflated incorrect dust structure in *NP-DOD-CAL*.

RC: ***– L58: Rephrase "fraud assumption" with false or incorrect assumption***

AR: Thanks for the comment. The "fraud" has been replaced as "incorrect" across the manuscript.

RC: ***– The Conclusions section would be strengthened by some discussion of the physical or statistical reasons why assimilation positively impacts certain vertical profiles while negatively impacting others***

AR: Thanks for the comment. We have added more discussions about the statistical reasons in the conclusion.

In conclusion, integrating ground- and satellite-derived aerosol observations into models enhances both the analysis and forecasting accuracy of the model's state. However, challenges persist in reconciling these observations with the model's high-dimensional state. Specifically, the model's initial, potentially flawed, vertical aerosol structure could impair assimilation efforts. The underlying reason is that data assimilation relies on the background error covariance to propagate the innovations between states and observations across the entire domain. If the vertical structure is inaccurate, this error may not only persist but could also be amplified during the assimilation process. Analytical examples from both ground-based and satellite-based assimilation confirm this effect. This paper has only scratched the surface by presenting a handful of negative instances, yet it is evident that this issue is pervasive in aerosol data assimilation. The path forward entails establishing a complementary network of vertical observations and implementing advanced assimilation methodologies that are sensitive to vertical structures, thereby offering a promising avenue to surmount these challenges.

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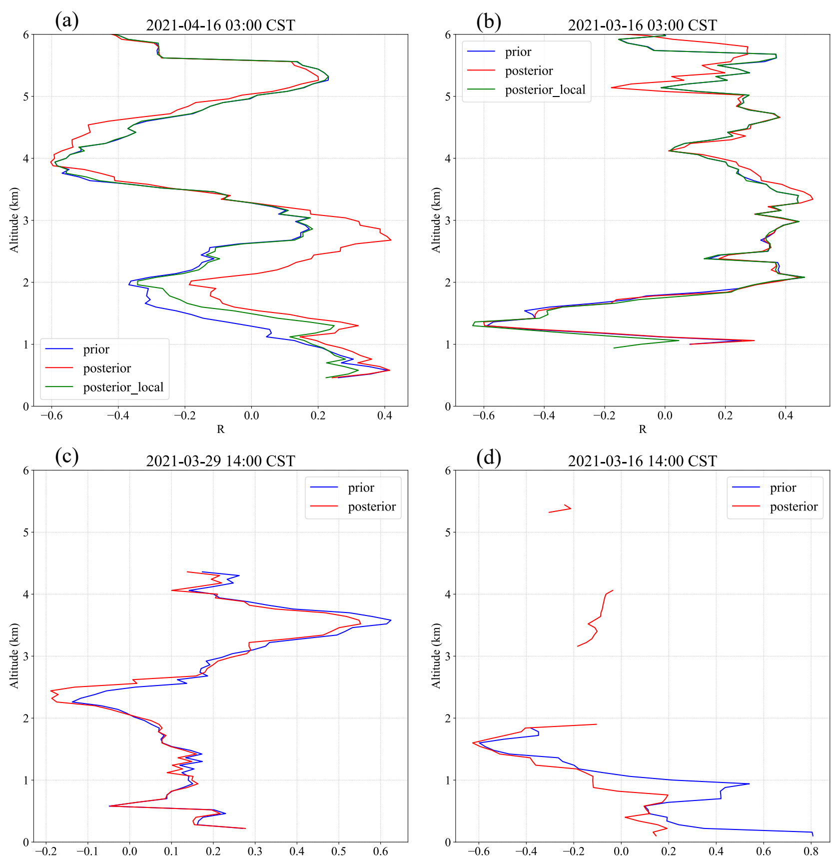

Figure 1: The correlation coefficient averaged on different altitudes. Each altitude contains all the paired data points (Concentration and extinction coefficient) along with the CALIPSO scan line

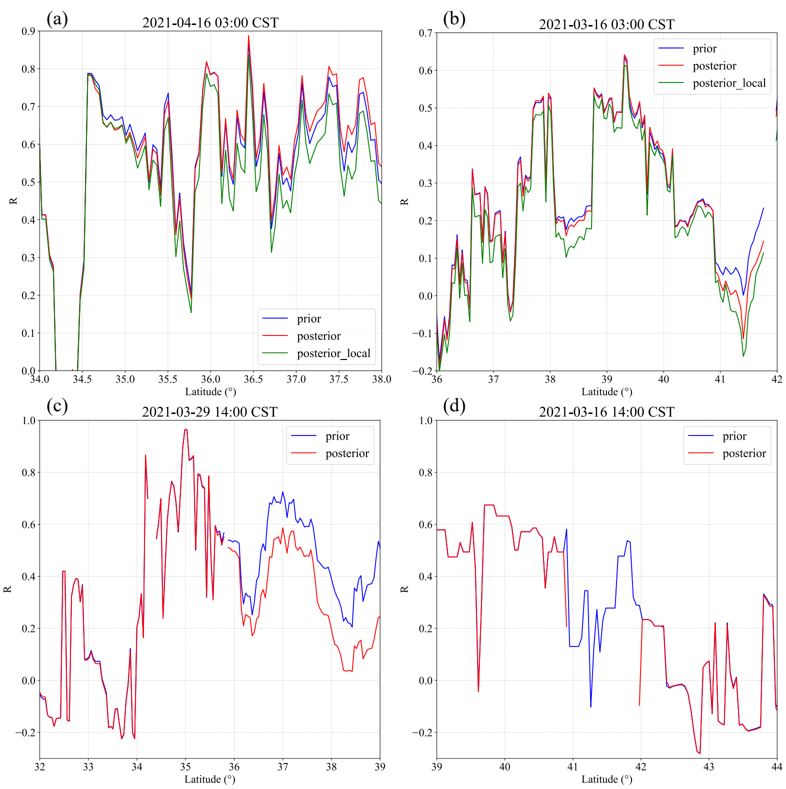

Figure 2: The correlation coefficient averaged on different latitudes. Each latitude contains all the paired data points (Concentration and extinction coefficient) along with the CALIPSO scan line

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