Response to Reviewer #1 for Geoscientific Model Development: Manuscript gmd-2024-148 By Liu et al.

We sincerely thank Reviewer #1 for thoughtful and constructive feedback. We have carefully considered each comment and made every effort to implement all the suggested changes. The notes below address each comment in detail. Please note that Reviewer's comments are shown in bold type and our responses in plain type.

Reviewer #1

General Comments:

This manuscript is the first part of a two-part paper that documents the development of a strongly coupled aerosol-meteorological 4D-Var assimilation system with part I focusing on the system description. This paper lands on an important topic, coupled data assimilation, which has gained more and more attention as the general consensus is to consider different components of the Earth system as a whole. Although the structure of the paper is quite well organized and the topic is very relevant to the geoscientific modeling community, I find the current form of the paper difficult to understand in two aspects: 1) English writing and 2) descriptive but lacks interpretation. For 1), there are many spelling errors, grammar errors, and inadequate use of words. In addition, the writing is unclear to an extent that I am unsure whether my interpretation of the concepts addressed in the paper is correct. For 2), it is important for a research paper to provide a thorough description of the result as well as to provide interpretation of the result (what does it imply, what could have caused the result, or does it make sense or not, etc). This paper did a good job at the descriptive part, but lacks interpretation, especially when discussing results from the single observation experiments and the real case experiments. With that, I recommend major revisions with many comments and questions listed below.

Response: We sincerely appreciate the reviewer's constructive feedback and the recognition of the importance of our study. We acknowledge the concerns regarding the clarity of the writing and the need for more interpretation of the results.

(1) English Writing: We have carefully reviewed and thoroughly revised the manuscript to improve the clarity, grammar, and overall readability. We have corrected spelling and grammatical errors, refined word choices, and restructured sentences where necessary to enhance the coherence of the text.

(2) Interpretation of Results: We appreciate the reviewer's suggestion to provide more interpretation of our findings. In the revised manuscript, we have significantly expanded the discussion of the results, particularly in the sections on single observation experiments and real case experiments.

We acknowledge that the original expression in Section 4.4 was not sufficiently clear, which may have caused confusion. We apologize for any misunderstanding. After carefully considering the reviewer's feedback, along with comments from the other two reviewers, we have completely rewritten this section, which is now presented as Section 5.3 in the revised manuscript.

In the updated version, we have clearly introduced the objective of the four experiments, which is to investigate the impact of different BC assimilation strategies on both BC and atmospheric variables. We have renamed the four experiments as DA_BC, DA_MET, DA_MET_then_BC, and DA_MET_BC_simult. The revised Table 3 now provides a clear description of the four experiments. We have also compared the BC analysis increments obtained from the DA_BC, DA_MET_then_BC, and DA_MET_BC_simult experiments, noting that the BC analysis increments from the DA_MET experiment are very small. Additionally, we compare the atmospheric analysis increments caused by BC assimilation in DA_BC, DA_MET_then_BC (DA_MET_then_BC - DA_MET), and DA_MET_BC_simult (DA_MET_BC_simult - DA_MET).

Our main conclusions from this analysis are as follows: The preliminary results obtained from this set of four experiments indicate that different BC assimilation strategies have little impact on BC analysis increments but significantly affect the analysis increments of atmospheric variables. When only BC observations are assimilated, the influence of BC on atmospheric variables is more pronounced, whereas the simultaneous assimilation of meteorological observations moderates this influence. This suggests that in BC assimilation, meteorological observations can help constrain the uncertainty introduced by BC observations on atmospheric variables, thereby improving the reliability of the assimilation results. Moreover, these results demonstrate the successful implementation of the newly developed CMA-GFS-AERO 4D-Var system and highlight it as an effective approach for investigating the feedback of BC data assimilation on meteorological forecasts.

In the future, we will conduct batch experiments using CMA-GFS-AERO 4D-Var to gain deeper insights into the role of BC assimilation in numerical weather prediction and further refine the system for broader applications.

Additionally, in response to Comment #24, we have adjusted the radius of influence for BC observations to 2 km, 10 km, and 20 km for urban, rural, and remote stations, respectively, according to Elbern et al. (2007). Consequently, all experiments in Section 5.3 have been redone using the updated radii, and the corresponding figures and text have been revised accordingly to reflect the new results.

For more details on the analysis, please refer to Section 5.3 of the revised manuscript. We once again appreciate the reviewer's valuable suggestions.

1. **As stated earlier, the writing is quite difficult to comprehend, preventing the readers from understanding the many seemingly important concepts and the value of the paper as a whole. Here are a few major concerns:**

   Response: Thanks for the valuable feedback. We sincerely apologize for the confusion caused by unclear writing in the manuscript. We have carefully revised the manuscript to improve its clarity and readability. We truly appreciate the reviewer's thoughtful suggestions for enhancing the quality of our manuscript.

   I. **Too many acronyms are used without being introduced at all. I only listed a few here: AURA/MLS, ARPEGE/MOCAGE, MOZART3, TM5, EUARD, IMAGES, STEM-III, CAMx, CMAQ, GEOS-Chem, GRAPES-CUACE, etc. Please make sure to introduce them when they were first mentioned and pay attention when referring to them at a later time.**

Response: Thanks for pointing this out. We have carefully reviewed the manuscript and ensured that all acronyms, including AURA/MLS (Microwave Limb Sounder aboard the Aura satellite), ARPEGE/MOCAGE (Action de Recherche Petite Echelle Grande Echelle/Modèle de Chimie Atmosphérique de Grande Echelle), MOZART3 (Model for Ozone and Related Chemical Tracers, version 3), TM5 (Transport Model, version 5), EUARD (The University of Cologne European Air Pollution Dispersion Chemistry Transport Model), IMAGES (Intermediate Model of Global Evolution of Species), STEM-III (Sulfur Transport Eulerian Model), CAMx (Comprehensive Air Quality Model with Extensions model), CMAQ (Community Multiscale Air Quality model), GRAPES-CUACE (Global/Regional Assimilation and PrEdiction System coupled with CMA Unified Atmospheric Chemistry Environmental Forecasting System), etc., are properly introduced when they are first mentioned.

GEOS-Chem is a global 3-D model of atmospheric chemistry driven by meteorological input from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office. CHIMERE is a three-dimensional chemical transport model used for atmospheric chemistry and air quality simulations. It was initially developed by the Pierre-Simon Laplace Institute (IPSL) in Paris, France. In the scientific literature, GEOS-Chem and CHIMERE are commonly referred to by their acronym, and there is no widely accepted or official full form for them. Therefore, we have followed the standard practice of using the acronym, consistent with other studies. Interested readers can refer to the relevant references in the manuscript for more detailed information.

Additionally, we have also ensured consistency and clarity when referring to them later in the text to avoid any confusion for the readers.

II. The word "field" is spelled incorrectly as "filed" in many places. Also, background field error covariance can be shortened to background error covariance. Furthermore, "feedback" does not always have to go with "effect".

Response: We sincerely apologize for the misspelling of the word "field", and this has been revised throughout the manuscript. Additionally, we have shortened "background field error covariance" to "<u>background error covariance</u>" as suggested. And "feedback effect" has also been revised to "<u>feedback</u>".

III. Inadequate use of word: the most frequently mis-used word is "set" in this paper, often times causing confusion and misunderstanding. For example, "the observation is set at 0300 UTC". A more accurate way could be "the observation is placed at 0300 UTC" or "the observation is assumed to take place at 0300 UTC". Here are a few more examples of imprecise use of word: "we set five experiments", "we set the single-point observation ideal experiment for BC", and "we further set the full observation experiment", etc.

Response: We sincerely apologize for the inadequate and imprecise use of the word "set", and we have revised "the observation is set at …" to "<u>the observation is placed at</u> …" as suggested. Additionally, we have revised "we set five experiments", "we set the single-point observation ideal experiment for BC", and "we further set the full observation experiment" to "we conducted five experiments", "we performed the single observation experiment for BC", "we further conducted the full observation experiment", respectively.

IV. **There is nothing wrong with calling it observation "increment", but a more appropriate term is observation "innovation". Also, I believe "single observation experiment" is a common term in data assimilation and there is no need to press on its "idealized" part.**

Response: According to the reviewer's good instructions, we have revised "observation increment" to "observation innovation". And we have also changed "single-point observation ideal experiment" to "single observation experiment" as suggested.

V. **There are many grammar errors and sentences that don't quite make sense. See comments below.**

Response: Thanks for the valuable feedback. We sincerely apologize for the grammar errors and unclear sentences in the manuscript. We have carefully reviewed the comments below and revised the manuscript to improve both clarity and grammatical accuracy. Specifically, we have addressed the highlighted issues and ensured that all sentences are well-structured and easy to understand. We truly appreciate the reviewer's attention to these details and are committed to enhancing the quality of the manuscript.

2. **Abstract: I am not against calling it a *chemistry* meteorology coupled data assimilation system, however, it makes more sense and less misleading to call it an *aerosol* meteorology coupled one since only black carbon (BC) aerosol is considered so far. Besides, the name of the system, CMA-GFS-AERO, actually already suggests that it is an aerosol-meteorology coupled system. Otherwise, it would be called CMA-GFS-Chem.**

Response: Thanks for the insightful comment. Following the recommendation, we have changed "chemistry meteorology coupled data assimilation system" to "aerosol-meteorology coupled data assimilation system" in Abstract and the main text.

3. **Introduction: While it makes sense to review the previous efforts on CCMM data assimilation focusing on the variational perspective, given that this study uses a 4DVar approach, it is also important to address the previous efforts on coupled aerosol-atmosphere data assimilation using the ensemble-based approaches. I suggest shortening the description on variational approach in the introduction and include some description of the ensemble approaches and highlighting the pros and cons of a variational choice relative to an ensemble approach. Obviously 4DVar is used since it's part of the CMA-GFS, it makes sense to extend upon the CMA-GFS 4DVar framework for aerosol coupling. Nevertheless, it is important to point out to the readers what to expect from coupling under a variational setup as opposed to an ensemble approach. For example, in a variational setup, the modeling of cross-variable component in background error covariance could be difficult, especially for aerosol vs. atmospheric processes, while in an ensemble setup one relies on ensemble estimation for**

cross-variable correlations. On the other hand, in a variational setup, the TLM and ADM are essential, and this can serve a natural transition to the next paragraph on the importance of ADM starting at line 52.

Response: Thanks for the insightful comment. Following the recommendation, we have shortened the description on variational approach in the introduction and included previous efforts on coupled aerosol-atmosphere data assimilation using the Ensemble Kalman filter (EnKF) method (Pagowski and Grell, 2012; Bocquet et al., 2015). We have also discussed the advantages and disadvantages of the 4D-Var method relative to the EnKF approach. Specifically, we have highlighted that the EnKF approach relies on ensemble-based estimates for the background error covariance, while in a variational setup, modeling cross-variable components in the background error covariance can be challenging in data assimilation for CCMM. Additionally, we have emphasized that in a 4D-Var framework, the TLM and ADM are essential, which naturally leads into the subsequent discussion on the significant advancements in atmospheric chemistry adjoint modeling.

We have updated the corresponding section of the Introduction as follows:

"…Flemming et al. (2011) utilized the 4D-Var system of the Integrated Forecast System (IFS) coupled with three different O_3 chemistry mechanisms, including a linear chemistry, the MOZART3 (Model for Ozone and Related Chemical Tracers, version 3) chemistry, and the TM5 (Transport Model, version 5) chemistry, to assimilate O_3 data from four satellite-borne sensors to improve the simulation of the stratospheric O_3 hole in 2008. Previous efforts have also explored the application of ensemble-based methods for data assimilation with a CCMM (Pagowski and Grell, 2012; Bocquet et al., 2015). Pagowski and Grell (2012) assimilated surface measurements of fine aerosols using the Weather Research and Forecasting-Chemistry model (WRF-Chem) and the Ensemble Kalman filter (EnKF) method. Bocquet et al. (2015) also presented an application of the EnKF to assimilate surface fine particulate matter observations and meteorological observations with the WRF-Chem model over the eastern part of North America. Results demonstrated that a large positive impact of aerosol data assimilation on aerosol concentrations, while the effect of meteorological observation assimilation on aerosol concentration is rather minor. All the preceding studies have laid good foundations for data assimilation with CCMM. However, since CCMM are fairly recent, the development and applications of data assimilation in CCMM are still limited. Further research and more attention are required, especially in terms of the potential feedbacks of chemical data assimilation on meteorological forecasts. Additionally, EnKF estimates background error covariance through ensemble forecasts, which rely on a limited number of ensemble members (Zhu et al., 2022). In high-dimensional problems, the limited number of samples may not be able to fully capture all the error characteristics, resulting the inaccurate of the estimation of background error covariance. In contrast, 4D-Var generally offers higher accuracy for high-dimensional problems by incorporating both the full observational data and model dynamics within the assimilation window, resulting in more precise state estimation. While the flow dependence of the background error covariance is implicitly realized within the assimilation window in 4D-Var, modeling the cross-variable component of the covariance presents a significant challenge in data assimilation for CCMM. Furthermore, the tangent linear model (TLM) and the adjoint model (ADM) are essential components of 4D-Var, but their development is often fraught with difficulties.

Significant efforts have been made in the field of atmospheric chemistry adjoint modeling. Elbern and Schmidt (1999) first constructed the ADM of a 3D CTM, EUARD (The University of Cologne European Air Pollution Dispersion Chemistry Transport Model). Inspired by this work…"

4. **Sections 3.1 & 3.2: It reads like there is a bunch of processes, programs, subroutines, and interfaces, but how they all work together to fulfill a coupled system is unclear. Please consider re-organize/re-write these two sections to increase clarity and make sure to stay consistent with what is being shown in Figs. S2 and S3. The key is to address the main processes in AERO-BC and describe what each process does. With that, it would make the readers easier to follow the subsequent TLM and ADM of AERO-BC section since all the pieces are there in the forward section already. In addition, it is not very clear what the interfaces that connect CMA-GFS with AERO-BC in all three model components (forward, TLM, and ADM) actually do in terms of coupling, other than knowing that they act to "couple" the aerosol with the atmosphere.**

Response: Thanks for the insightful comment. Following the recommendation, we have rewritten Sections 3.1 and 3.2 to improve clarity. In doing so, we also carefully considered Comment #16 and Comment #17 and concluded that Figures S2 and S3 were not essential. The revised Sections 3.1 and 3.2 now provide a clearer and more self-contained description of the key processes in AERO-BC, making the figures unnecessary. The revised Sections 3.1 and 3.2 explicitly describe the key processes in AERO-BC and their respective roles. Additionally, we have clarified the function of the interface programs that connect CMA-GFS with AERO-BC in all three model components (forward model, TLM, and ADM), ensuring a clearer explanation of their coupling mechanism. These interface programs are responsible for transferring meteorological parameters (e.g., temperature, wind, and humidity) from CMA-GFS to AERO-BC, extending the spatial dimension from 1-D to 3-D, and reading emissions for AERO-BC.

The revised Sections 3.1 and 3.2 now reads:

"3.1 CMA-GFS-AERO forward model

In this work, for the sake of interest in BC and the consideration of computational efficiency, we developed the CMA-GFS-AERO forward model by integrating the aerosol module AERO-BC into CMA-GFS v4.0. The AERO-BC module was created by extracting BC-related codes from the CUACE model, with its functionality aligning with the BC aerosol processes in the CAM module of CUACE. In other words, the physical processes for BC in AERO-BC are identical to those in the CAM module, with no changes made. The main differences lie in the engineering aspect: (1) while the CAM module was originally written in Fortran 77, the AERO-BC code has been rewritten in Fortran 90; (2) since CAM in CUACE deals with six types of aerosols, the code structure is somewhat complex and redundant, whereas AERO-BC focuses solely on BC, resulting in a simpler and more streamlined structure. These updates improve code readability and enhance computational efficiency, without affecting the underlying physical processes.

…

The main processes in AERO-BC include: (1) calculating the emission flux of BC through the surface flux calculation module, (2) calculating the vertical diffusion trend of BC by solving the vertical diffusion equation, and (3) simulating key BC aerosol processes in the atmosphere, including hygroscopic growth, coagulation, nucleation, condensation, dry deposition/sedimentation, and below-cloud scavenging. For more details, please refer to the relevant literature on the CAM module (Gong et al., 2003; Gong and Zhang et al., 2008; Wang et al., 2010; Zhou et al., 2012). In the integration of AERO-BC with CMA-GFS, the interface programs transfer meteorological parameters (e.g., temperature, wind, and humidity) from CMA-GFS to AERO-BC, extend the spatial dimension from 1-D to 3-D, and read emissions for AERO-BC. The transport processes for ψ_{bc}^{n} are the same as those for the variables associated with the different water species in CMA-GFS, using the hybrid PRM and QMSL schemes (Su et al., 2013).

…

3.2 CMA-GFS-AERO TLM and ADM

…

The TL of the AERO-BC can be obtained by linearizing $\boldsymbol{F}$, expressed as

$$\delta Y = \mathbf{F}\delta C = \frac{\partial \boldsymbol{F}}{\partial C}\delta C, \quad (5)$$

where $\mathbf{F}$ is the TL model operator, δC and δY represent perturbations of input and output variables of the AERO-BC, respectively.

The adjoint of the AERO-BC is essentially the transpose of the AERO-BC TL, expressed as

$$\delta C^{*} = \mathbf{F}^{T}\delta Y^{*}, \quad (6)$$

where $\mathbf{F}^{T}$ is the adjoint operator of $\mathbf{F}$, δY^{*} and δC^{*} represent input and output variables of the adjoint of AERO-BC, respectively.

In constructing the TL and the adjoint of AERO-BC, no simplifications were made to the AERO-BC processes. Specifically, no regularization was applied to the nonlinear equations, nor were any complex processes, which were difficult to linearize, omitted. As a result, the TL and the adjoint of AERO-BC fully include all processes related to emission flux, vertical diffusion, and aerosol physical processes as described in Section 3.1.

The TL and the adjoint of AERO-BC are 1-D modules with fixed latitude and longitude coordinates. To extend them to 3-D, the tangent linear and the adjoint of the interface programs were also constructed. Furthermore, the tangent linear and the adjoint of BC transport processes follow the same framework as those for the variables associated with the different water species in the CMA-GFS TLM and ADM, utilizing the tangent linear and the adjoint of QMSL. In this way, the 3-D parameters could be transferred from CMA-GFS to AERO-BC. Thus, we obtained the CMA-GFS-AERO TLM and ADM."

5. **"Section 4.1 Model setup" should be separated from the Result section since it is not a result but a description of model configuration or model setup. It might be better to consider it as a standalone section or to be included as a sub-section of section 3.**

Response: We sincerely appreciate the reviewer's valuable suggestion. Following the recommendation, we have separated the "Model setup" from the Results section and designated it as a standalone section, now titled "4 Model setup". Consequently, the Results and Conclusions sections have been renumbered as Section 5 and 6, respectively. Additionally, we have updated the corresponding descriptions in the Introduction as follows:

"Section 2 introduces the methods, Section 3 describes the development of CMA-GFS-AERO

4D-Var, Section 4 provides the model setup, Section 5 presents the results, and the conclusions are found in Section 6."

6. **Page 2, Lines 32-34: What exactly are these moisture and temperature perturbations? And what these perturbations to dynamics?**
   Response: Thanks for the valuable comment. The "moisture and temperature perturbations" refer to changes in the moisture and temperature fields in the atmosphere that result from aerosol microphysics and radiative forcing. These changes occur due to the interactions between aerosols, radiation, and cloud processes, which alter the local moisture and temperature distributions. The "perturbations to dynamics" refer to how these changes in moisture and temperature affect atmospheric processes such as convection, circulation, and stability, which in turn influence the overall atmospheric dynamics. This feedback mechanism is incorporated in the coupled chemistry meteorology models (CCMM), but is not typically included in chemical transport models (CTM).
   To enhance clarity, we have revised lines 32-34 as follows:
   "…CCMM account for the feedback mechanism between aerosols and meteorology, specifically the moisture and temperature perturbations resulting from aerosol microphysics and radiative forcing, which, in turn, affect atmospheric dynamics such as convection, circulation, and stability, whereas CTM lack the capability to incorporate these feedback mechanisms ..."

7. **Page 2, Line 35: "enabling to produce the optimal initial values for …" > "enabling the production of an optimal initial condition for …"**
   Response: This has been revised as suggested.

8. **Page 2, Line 52: I am not sure whether "international mainstream" is a good way to say it here. How about just "major"? Also, it should be "numerical weather prediction centers", not "numerical weather centers".**
   Response: Thanks for pointing this out. We agree that "international mainstream" is not a good way to say it here, and "major" is a more appropriate term. We also agree that it should be "numerical weather prediction centers", not "numerical weather centers". Based on the third suggestion in the previous comment, we have revised this section and removed the sentence entirely.

9. **Page 3, Lines 72-74: only the surface temperature? What happened to the 3D temperature field?**
   Response: Thanks for the insightful comment. The impact of black carbon (BC) on temperature is indeed not limited to the surface. BC influences both the surface temperature and the three-dimensional (3D) temperature field through its absorption of solar radiation in the visible to infrared wavelength range. We have revised Lines 72-74 as follows:
   "…BC is also the main optically absorbing component of atmospheric aerosols, effectively absorbing solar radiation in the visible to infrared wavelength range, thus affecting not only the surface temperature but also the 3D temperature field…"

10. Page 3, Line 86 & Page 8, Line 211: "adding the control variable of BC into …" > "adding BC as a control variable into …"

Response: This has been revised as suggested.

11. Page 4, Lines 103-104: I am not sure what is meant by "freely combinable"? These physical parameterization processes are common to many global models. What is more important is which "schemes" are being used in each of these physical processes in CMA-GFS.

Response: Thanks for the insightful comment. By "freely combinable", we are referring to the flexibility in choosing among several physical parameterization schemes for a specific physical process, allowing users to select the most suitable one for their needs.

According to the reviewer's valuable suggestions, we have modified the expression to clarify the specific schemes used for each physical process in CMA-GFS in this study, as follows:

"…The physical parameterization schemes used in this work mainly include the Simplified Arakawa Schubert (SAS) cumulus convection scheme (Arakawa and Schubert, 1974; Liu et al., 2015), the double-moment cloud microphysics scheme (Liu et al., 2003a, 2003b; Li et al., 2024), the Rapid Radiative Transfer Model for the GCM (RRTMG) longwave and shortwave radiation schemes (Mlawer et al., 1997; Morcrette et al., 2008), the Common Land Model (CoLM) land surface scheme (Dai et al., 2003), and the New Medium Range Forecast (NMRF) boundary layer scheme (Hong and Pan, 1996; Han and Pan, 2011)…"

12. Page 4, Line 114: what is sectional representation method? And is there a reference for that?

Response: Thanks for pointing this out. Sectional representation is one of the common methods to represent particle size distributions in atmospheric chemistry models. In the sectional representation approach, the aerosol size distribution is generally approximated by a set of contiguous, nonoverlapping and discrete size bins. This representation of aerosol size distribution is employed for its flexibility to treat processes including multicomponent interactions such as coagulation, condensation and chemical processes. We list several references related to the sectional representation method here:

[1] Gelbard, F., Tambour, Y., Seinfeld, J. H.: Sectional representations for simulating aerosol dynamics. J. Colloid Interf. Sci., 76, 541-556, https://doi.org/10.1016/0021-9797(80)90394-X, 1980.

[2] Meng, Z., Dabdub, D., Seinfeld, J. H.: Size-resolved and chemically resolved model of atmospheric aerosol dynamics, J. Geophys. Res., 103, 3419-3435, https://doi.org/10.1029/97JD02796, 1998.

[3] Gong, S.L., Barrie, L.A., Blanchet, J.P., Von Salzen, K., Lohmann, U., Lesins, G., Spacek, L., Zhang, L.M., Girard, E., Lin, H.: Canadian Aerosol Module: A size - segregated simulation of atmospheric aerosol processes for climate and air quality models 1. Module development, J. Geophys. Res.-Atmos., 108, AAC 3-1-AAC 3-16, https://doi.org/10.1029/2001JD002002, 2003.

We have also added an explanation and the references of sectional representation method in the revised manuscript as follows:

"…and each of them utilizes the sectional representation method (Gelbard et al., 1980; Meng et al., 1998; Gong et al., 2003), in which the aerosol size distribution is generally approximated by a set of contiguous, nonoverlapping and discrete size bins, to represent particle size distributions…”

13. **Page 5, Lines 125: 137: M0>i and M^T0>i are actually linear and adjoint “models”, not “operators”.**
    Response: This has been revised as suggested.

14. **Page 5, Lines 134-135: “after the physical and preconditioning transformation” can be omitted since it has already been stated in line 132.**
    Response: This has been revised as suggested.

15. **Page 6, Line 159: To be consistent with the wordings at line 155, please consider using “forward model” instead of CCMM.**
    Response: According to the reviewer’s good instructions, we have used “CMA-GFS-AERO forward model” instead of “CMA-GFS-AERO CCMM” throughout the manuscript.

16. **Page 6, Lines 160-164: These are not very relevant information.**
    Response: Thanks for the comment. The content in Lines 160-164 has been rewritten in the revised manuscript. For details, please refer to our response to Comment #4.

17. **Page 6, Lines 163-165 and Figure S2: These descriptions are not consistent with what is shown in Fig. S2 (a) and (b). If the idea is to show that bc_driver is part of the CMA-GFS-AERO model and acts as the interface of AERO-BC to CMA-GFS, if can be simply stated without showing Fig. S2a. As for Fig. S2b, while sf_bc, trac_vert_diff, and aerosol_bc are listed, the constant/parameter program (as stated in the texts) is missing. If the subroutines under each program is important for the readers to know and will be used/mentioned in the later part of the paper, then they deserve some explanation (e.g., what is cal_aerosol_prop? some sort of calculation of aerosol optical properties?), otherwise, they need not to be mentioned or shown. For example, the q2rh program seems to be irrelevant to AERO-BC, perhaps it can be omitted to help the readers put their focus on only the relevant parts.**
    Response: Thanks for the detailed comments. In response to both Comment #4 and Comment #16, we have rewritten Sections 3.1 and 3.2. The revised Sections 3.1 and 3.2 now provide a clearer and more self-contained description of the key processes in AERO-BC, making the figures unnecessary. Therefore, we have removed Figures S2 and S3 from the manuscript. For more details, please refer to our response to Comment #4.

18. **Page 6, Line 172: it makes more sense to mention the index for size bin of BC here, instead of later at section 3.3.1, as the idea of 6 diameter bins is introduced here: Ψbc > Ψbcn where n = 1, 6.**
    Response: We sincerely appreciate the reviewer’s valuable suggestion. Following the recommendation, we have introduced the index for the size bin of BC in Section 3.1 as follows: “Thus, six new prognostic variables for the mass mixing ratio of BC, denoted as ψ_{bc}^{n}(unit: kg/kg),

where $n = 1, ..., 6,$ are added in the dynamical framework of CMA-GFS.”

Accordingly, we have removed the introduction of ψ_{bc}^{n} in Section 3.3.1. Additionally, all occurrences of ψ_{bc} throughout the manuscript have been updated to ψ_{bc}^{n}.

19. **Page 6, Line 173: “water-matter variables”: are these water vapor and hydrometeor habits mass mixing ratios?**

Response: Thanks for the comment. We sincerely apologize for the lack of clarity regarding the term “water-matter variables”. By “water-matter variables”, we were referring to the variables associated with the different water species. In the revised manuscript, we have replaced “water-matter variables” with “variables associated with the different water species” for clarity.

20. **Page 7, Lines 183-184: this last sentence about TLM and ADM codes being written line-by-line manually doesn’t seem quite necessary. Why is it important to mention that the code is written manually without using any automatic differentiation tool?**

Response: Thanks for pointing this out. Zou et al. (1997) emphasized that due to the complexity of numerical model codes, even when independent, dependent, and active variables are correctly specified, automatic adjoint generators do not necessarily produce correct adjoint codes. Clean and accurate adjoint codes often require manual intervention. Our experience in developing the CMA-GFS adjoint model (Zhang et al., 2019) further confirms that adjoint codes generated by automatic differentiation tools often suffer from issues such as poor readability, poor maintainability, low efficiency, and even errors. To ensure the quality, readability, maintainability, and efficiency of the tangent linear and adjoint codes, we opted to write them line-by-line manually, without using any automatic differentiation tool.

We have also added an explanation in the revised manuscript as follows:

“…Since adjoint codes generated by automatic differentiation tools often suffer from issues such as poor readability and maintainability, low efficiency and even errors due to the complexity of numerical models (Zou et al., 1997), the tangent linear and adjoint codes in this study were written line-by-line manually, without using any automatic differentiation tool.”

21. **Page 8, Lines 231-233: does this suggest that distribution weight only depends on the size bin, and does not vary spatially? meaning that all grid points use the same distribution weight for a given size bin? If so, is it guaranteed that BC mass conserved after the re-distribution?**

Response: Thanks for the insightful comment. In our methodology, the distribution weights (ω^n) are calculated based on the entire three-dimensional domain, following the equation $\omega^n = \frac{\sum_1^N {\psi_{bc}}^n}{\sum_{n=1}^6 \left(\sum_1^N {\psi_{bc}}^n\right)}$, where N represents the number of three-dimensional grid points. This means that for a given size bin, ω^n is uniform across all grid points and does not vary spatially. This approach ensures that the weight distribution reflects the global characteristics, rather than being influenced by local variations. By doing so, a global weighting factor is provided, which allows for a reasonable allocation of analysis increments. While there may be small variations in the distribution weights (ω^n) across different grid points, these differences are relatively minor. Therefore, using a global weighting factor does not result in a significant violation of BC mass conservation.

22. Page 8, Line 245: what is AE31?

Response: Thanks for pointing this out. The AE31 is a model of the Aethalometer manufactured by Magee Scientific (USA), which is widely used for the real-time measurement of BC concentration. The AE31 determines the mass concentration of BC particles collected from air samples, flowing through a quartz filter. The instrument measures the transmission through the filter over a wide spectrum of wavelengths from 370 nm to 950 nm. Light at the selected wavelength is transmitted through control and sample filters, and the attenuation change in the filter is then translated into the BC mass concentration. In our study, we used the BC concentration measured at the recommended wavelength of 880 nm.

We have revised the description of AE31 in Section 3.3.2 as follows:

"…The monitoring of BC in CAWNET was conducted using an Aethalometer, AE31, which is one of the models produced by Magee Scientific (USA, https://www.aerosolmageesci.com). The AE31 determines mass concentration of BC particles collected from air samples, flowing through a quartz filter. The instrument measures the transmission through the filter over a wide spectrum of wavelengths from 370 nm to 950 nm. Light at the selected wavelength is transmitted through control and sample filters, and the attenuation change in the filter is then translated into the BC mass concentration. In this study, we used the BC concentration measured at the recommended wavelength of 880 nm…"

23. Page 9, Lines 246-247: what are the quality control procedures?

Response: Thanks for pointing this out. The original sampling frequency of the AE31 is 5 minutes. Due to various factors, including instrument-related issues or human error, the BC observations may contain abnormal values such as missing values, negative values, or extreme outliers. Therefore, quality control is necessary before using the BC concentrations in our analysis. In this study, the quality control procedures for BC observations mainly focus on eliminating abnormal values and filling in missing data. The quality control steps are as follows:

(1) Eliminating abnormal values. During the calculation of hourly average values from the 5-minute sampled data, any BC concentration values that are significantly different from the hourly average (i.e., those where the absolute difference exceeds three times the standard deviation) are considered abnormal and discarded. Additionally, any bad data flagged by the instrument's monitoring system are also removed.

(2) Filling in missing values. If more than one-third of the data for a given hour is missing, or if there are more than three consecutive missing values, the entire hour's data is discarded. For other cases, linear interpolation is applied to fill in the missing values.

After applying these quality control procedures, we obtained the hourly average BC concentrations used in this study.

We have added detailed quality control procedures in Section 3.3.2 as follows:

"…The AE31 measures BC concentrations every 5 minutes. We performed quality control on the original data and obtained the hourly average values, which were used in the BC assimilation experiments. The quality control procedures are as follows:

(1) Eliminating abnormal values. During the calculation of hourly averages from the 5-minute

sampled data, any BC concentration values that differ significantly from the hourly average (i.e., those where the absolute difference exceeds three times the standard deviation) are considered abnormal and discarded. Additionally, any bad data flagged by the instrument's monitoring system are also removed.

(2) Filling in missing values. If more than one-third of the data for a given hour is missing, or if there are more than three consecutive missing values, the entire hour's data is discarded. For other cases, linear interpolation is applied to fill in the missing values."

24. **Page 9, Line 257: According to Table 3 of Elbern et al. (2007), the radius of influence varies with station types, and a radius of 10 km corresponds to a rural station. Since 10 km is selected here, does that mean all 32 CAWNET stations are all rural stations? If not, please provide justifications for using a radius of 10 km.**

    Response: Thanks for the insightful comment. The 32 CAWNET stations include 11 urban, 17 rural and 4 remote stations. As noted in Table 3 of Elbern et al. (2007), the radius of influence does vary with station types. Our initial selection of a uniform 10 km radius for all 32 CAWNET stations was indeed inappropriate. In the revised manuscript, we adopted radii of 2 km, 10 km, and 20 km for urban, rural, and remote stations, respectively, according to Table 3 of Elbern et al. (2007). The following revisions have been made in the text:

    "The BC observation data were collected from 32 stations (Guo et al., 2020), including 11 urban, 17 rural and 4 remote stations…"

    "…and L is the radius of influence of a BC observation. According to Elbern et al. (2007), L was set to 2 km, 10 km, and 20 km for urban, rural, and remote stations, respectively…"

    Additionally, all experiments in Section 5.3 have been redone using the updated radii, and the corresponding figures and text have been updated to reflect the new results. Please refer to the revised Section 5.3 for details.

25. **Page 9, Lines 264-268: I have trouble understanding this sentence… what is point jump and what does layer jump mean?**

    Response: We sincerely apologize for the unclear expression of "point jump" and "layer jump". What we intended to express is as follows:

    In a data assimilation system, the observation operator serves two primary functions: (1) transforming model state variables into observed physical quantities and (2) interpolating the background (or analysis) field to the observation locations. The transformation of physical quantities depends on the observation type, while the spatial interpolation consists of both horizontal and vertical components. Since the CMA-GFS-AERO 4D-Var system adopts the Charney-Phillips staggered grid in the vertical direction and the Arakawa-C grid in the horizontal direction, different physical variables (e.g., wind, temperature, etc.) are positioned at different locations within the model grid. To minimize errors introduced by variable transformations and spatial interpolation in the observation operator, it is necessary to correctly account for the staggered placement of variables. This involves handling the horizontal staggering of grid points and the vertical staggering of layers when interpolating observations.

    Additionally, we have revised the corresponding sentence in the manuscript as follows:

    "…Since the CMA-GFS-AERO 4D-Var system adopts the Charney-Phillips staggered grid in the vertical direction and the Arakawa-C grid in the horizontal direction, the observation operator must account for the staggered locations of different physical variables. To minimize errors introduced by variable transformations and spatial interpolation, appropriate handling of horizontal staggering and vertical layer transitions is required…"

26. **Page 10, Line 269: "accumulated" > "summed" ?**
    Response: This has been revised as suggested.

27. **Page 10, Lines 281-282: what is the physical meaning of such a simplification that assumes correlation coefficient is a product of vertical one times the horizontal one? What does this simplification imply?**
    Response: Thanks for the valuable comment. In the CMA-GFS 4D-Var assimilation system, the background error covariance matrix is highly complex due to the presence of both inter-variable correlations and correlations at different spatial locations of the same variable. Given the large dimensionality of the problem, direct computation is not feasible, and a dimensionality reduction approach is necessary.
    In the CMA-GFS 4D-Var system, the state variables (such as the non-dimensional pressure, potential temperature, horizontal wind components, vertical wind component, and specific humidity) are first transformed through physical transformations into the stream function, non-equilibrium velocity potential, non-equilibrium dimensionless pressure , and specific humidity. These variables are independent of each other, so only the error covariance between the same variables at different spatial locations exists. As a result, the background error covariance matrix becomes a block diagonal matrix.
    Further, through preconditioning transformations, the background error covariance matrix is simplified to an identity matrix. During this transformation process, we assume that the correlations at different spatial positions for the same variable can be separated into horizontal and vertical components. Specifically, the vertical correlation model is assumed to be identical in the horizontal direction, and the horizontal correlation model is assumed to be the same in the vertical direction. This allows us to represent the background error covariance matrix using the Kronecker product of the horizontal and vertical correlation matrices, significantly simplifying the computation of the matrix.
    For further details, please refer to Zhang et al. (2019): Zhang, L., Liu, Y., Liu, Y., Gong, J., Lu, H., Jin, Z., Tian, W., Liu, G., Zhou, B., Zhao, B.: The operational global four - dimensional variational data assimilation system at the China Meteorological Administration, Q. J. Roy. Meteor. Soc., 145, 1882-1896, https://doi.org/10.1002/qj.3533, 2019.
    We have also included this reference in the revised manuscript as follows:
    "…Therefore, in the CMA-GFS 4D-Var assimilation system, a simplification is made by assuming that the correlation coefficient can be expressed as the product of the vertical correlation coefficient and the horizontal correlation coefficient (Zhang et al., 2019)…"

28. **Page 10, Line 290: what does K_p represent and why set it to 10 here?**
    Response: Thanks for the comment. k_p is the constant coefficient in the formula. In this study, we set the value of k_p to 10 for the control variable BC, following the value used for the control variable of humidity in the CMA-GFS 4D-Var system.

Additionally, we have included an explanation for k_p in the revised manuscript as follows: "…and k_p is the constant coefficient (Bergman, 1979). Following the value of k_p used for the control variable of humidity in the CMA-GFS 4D-Var system, we set k_p to 10 for the control variable BC…"

29. **Page 11, Lines 301-302: "referenced to the relationship between length scale of humidity and the height": I have trouble understanding this one as well. Why is a relationship between humidity length scale and "height" being used for the "horizontal" length scale of BC?**
Response: Thanks for pointing this out. We sincerely apologize for the confusion caused by the unclear expression in Lines 301-302. What we intended to convey is that the length scale for the control variable BC varies with height in the model, following the way the length scale of the humidity variable varies with height in the CMA-GFS 4D-Var system.
We have revised the expression in Lines 301-302 as follows:
"…The length scale for the control variable BC varies with height in the model, following the way the length scale of the humidity variable varies with height in the CMA-GFS 4D-Var system, which is shown in Table 1…"

30. **Page 12, Lines 313-315: does this mean that BC is not cycled since the model is restarted every 6 h from CMA-GS analysis that does not have BC? But the next sentence seems to indicate that 6-h forecast of BC is used as background for the next cycle… these are conflicting ideas.**
Response: We sincerely apologize for the unclear expression in Lines 313-315, which may have caused confusion. To clarify, we have revised the text in the revised manuscript as follows:
"…The forecast of the CMA-GFS-AERO model started at 0300 UTC on October 1, 2016, and was restarted every 6 h. The meteorological initial fields for each 6-h cycle were obtained from the operational CMA-GFS analysis. The BC field was initialized with null concentrations at 0300 UTC on October 1, 2016. From the second forecast cycle onward, the initial conditions of BC were derived from the BC field at the end of the previous 6-h forecast, allowing the BC field to be cycled…"

31. **Page 12, Line 321: what does a global scale actually mean here? Resolution, data coverage, etc?**
Response: Thanks for pointing this out. By "a global scale", we are referring to the data coverage of the HTAP, which provides emissions data for a wide range of regions worldwide. We have modified the expression in the revised manuscript as follows:
"…and the global datasets of the Task Force Hemispheric Transport of Air Pollution (HTAP) (Janssens-Maenhout et al., 2015) …"

32. **Page 12, Line 329: "an important part of introducing an adjoint model" > "an important part of introducing a new modeling component, such as the AERO-BC module"?**
Response: This has been revised as suggested.

33. **Page 13, Lines 345-346: “in an approximately linear way” > “in an approximately linear manner”?**
Response: This has been revised as suggested.

34. **Page 14, Lines 377-378: 6-h integration seems a rather long time. Is it possible that the AERO-BC processes are not very nonlinear?**
Response: Thanks for the insightful comment. Since black carbon (BC) does not participate in complex chemical reactions, the AERO-BC processes in the CMA-GFS-AERO forward model primarily include emission flux, vertical diffusion, coagulation, nucleation, condensation, and dry deposition/sedimentation. As the reviewer correctly points out, these processes are treated as approximately linear in the model and do not exhibit strong nonlinear behavior. Consequently, a 6-hour integration time is reasonable and does not introduce significant errors due to process nonlinearity. Additionally, the assimilation time window of the CMA-GFS-AERO 4D-Var system is 6 hours. Using a 6-hour integration time aligns with this window and enables the evaluation of the tangent linear model (TLM) performance. Our results confirm that the CMA-GFS-AERO TLM demonstrates good performance in the tangent linear approximation of BC.

35. **Page 15, Lines 391-392: I have trouble understanding this one. Which coupled variable? And which physical process variable? Is it also possible that AERO-BC processes are not very nonlinear such that TL approximation is not too much different from the NL one?**
Response: We sincerely apologize for the lack of clarity in this sentence. By “coupled variable”, we are referring to BC, and “physical process variable” refers to variables such as specific humidity. What we want to explain is that compared with variables such as potential temperature and specific humidity in the CMA-GFS-AERO model, the tangent linear approximation for BC is quite effective, making it well-suited for constructing a 4D-Var system. We have revised the sentence in the manuscript for better clarity as follows:
“…This phenomenon indicates that, in comparison to variables such as potential temperature and specific humidity in the CMA-GFS-AERO model, the tangent linear approximation for BC is quite effective, making it well-suited for constructing a 4D-Var system.”
As the reviewer correctly points out, the AERO-BC processes are treated as approximately linear in the model and do not exhibit strong nonlinear behavior. As a result, TL approximation of the AERO-BC is not too much different from the NL one.

36. **Page 18, Lines 453-457: It will be quite helpful to add more texts to address the links between Fig. 5a and Fig. 6a as these two figures are results from the same single observation experiment with observation placed at the beginning of the window (i.e., 0300 UTC) where Fig. 5a shows the initial analysis increment while Fig. 6a shows the propagated analysis increment valid at the end of the window. Same idea for Fig. 5b and Fig. 6b, while the only difference is the timing of the observation.**
Response: Thanks for the valuable suggestion. We have added more explanations to clarify the links between Fig. 5 and Fig. 6 in the revised manuscript as follows:

"…For the case where the BC observation is placed at 0300 UTC, the initial analysis increment at 0300 UTC (Fig. 5a) exhibits an isotropic structure due to the static B. In contrast, the propagated analysis increment at the end of the assimilation time window (0900 UTC, Fig. 6a) exhibits an anisotropic structure under the influence of the flow-dependent $M_{0\to i}BM_{0\to i}^{T}$. Similarly, when the BC observation is placed at 0600 UTC, both the initial analysis increment at 0600 UTC (Fig. 5b) and the propagated analysis increment at 0900 UTC (Fig. 6b) exhibit an anisotropic structure. In addition, the horizontal distribution structure of the BC analysis increments in Fig. 6a and Fig. 6b closely resembles that of the analysis increments at the observation time of 0900 UTC (Fig. 5c). This indicates the significant impact of flow-dependent dynamics on the evolution of the analysis increments. No matter what time the observation is placed at, the spatial propagation of the observation information is effectively achieved through the model integration."

37. **Page 19, Lines 467-469: while I think I understand what the authors are trying to say, it is not entirely correct and perhaps not necessary to end the sentence like this. The way the system is setup (i.e., the CMA-GFS-AERO 4DVar system) by minimizing both BC and atmospheric variables together suggests it is a coupled assimilation. I think what the authors are trying to suggest is that the merits of a coupled data assimilation system cannot be fully manifested or exploited by only assimilating a BC observation at the beginning of the window.**

    Response: We sincerely thank the reviewer for the valuable comment. We agree with the reviewer's suggestion and have revised the sentence for clarity and accuracy. The original wording was indeed not entirely correct, and we appreciate the clarification regarding the coupled nature of the system. To better reflect the intended meaning, we have rephrased the sentence to emphasize that the full potential of a coupled data assimilation system is not realized by only assimilating a BC observation at the beginning of the assimilation window. The revised sentence now reads:

    "…In this case, the merits of a coupled data assimilation system cannot be fully manifested by only assimilating a BC observation at the beginning of the window…"

38. **Page 19, Lines 467-476: I think it is nice to have a paragraph detailing the processes in the 4DVar component of CMA-GFS-AERO that induces non-zero cross-covariance between the atmosphere and BC variables via evolving the initial background covariance with the TL modeling, even though the initial crosscovariance is zero. The current paragraph is trying to do so but remains rather descriptive and lacks interpretation. For that, I suggest checking out Section 2.1 "Coupled data assimilation" of Smith et al. (2015). Smith, P. J., Fowler, A. M., & Lawless, A. S. (2015). Exploring strategies for coupled 4D-Var data assimilation using an idealised atmosphere–ocean model. Tellus A: Dynamic Meteorology and Oceanography, 67(1). https://doi.org/10.3402/tellusa.v67.27025**

    In addition, I do not think "co-correlation" is a proper word.

    Response: Thanks for the insightful comment. We have referred to Section 2.1 "Coupled data assimilation" of Smith et al. (2015) as suggested and have added Section 3.3.5 in the revised manuscript to provide a more detailed interpretation of the mechanisms behind the evolution of the initial background covariance. The updated section reads as follows:

"3.3.5 Flow-dependent background error covariance in CMA-GFS-AERO 4D-Var
In the strongly coupled aerosol-meteorology assimilation system, interactions between the atmospheric variables and BC allow BC observations to influence the analysis increment of atmospheric variables and vice versa. The incremental 4D-Var algorithm implicitly evolves the background error covariances ($\mathbf{B}$) throughout the assimilation window according to the TL model dynamics. This process modifies prior background error variance estimates and induces non-zero correlations between model variables (Smith et al., 2015). By utilizing the fully coupled TLM and ADM in the inner loops of the strongly coupled assimilation system, cross-covariance information between BC and atmospheric variables is generated. This enables observations of one variable to produce analysis increments in the other, leading to more consistent analyses.

Specifically, if the BC observation is assumed to take place at the initial of the assimilation window, the 4D-Var assimilation is equivalent to the 3D-Var assimilation. Since the BC variable is assumed to be uncorrelated with the atmospheric variables in the static $\mathbf{B}$, and there is no direct relationship between the BC observation operator and the atmospheric variables, the BC observation does not lead to the generation of the analysis increments of atmospheric variables. In this case, the merits of a coupled data assimilation system cannot be fully manifested by only assimilating a BC observation at the beginning of the window. If the BC observation is assumed to take place at the middle and the end of the assimilation window, $\mathbf{B}$ evolves within the assimilation time window through the TLM $\mathbf{M}_{0\to i}$, obtaining the implicit background error covariance matrix $\mathbf{M}_{0\to i}\mathbf{B}\mathbf{M}_{0\to i}^{T}$ that evolves with time. $\mathbf{M}_{0\to i}\mathbf{B}\mathbf{M}_{0\to i}^{T}$ includes the cross-covariances information of BC and atmospheric variables, and can realize the feedback of the BC observation to the atmospheric variables through the CMA-GFS-AERO ADM $\mathbf{M}_{0\to i}^{T}$, further producing analysis increments of atmospheric variables."

Additionally, we have revised the description of Figure 7 in the manuscript to make it more concise:

"Figure 7 depicts the analysis increments of temperature at the first model level at the initial time of the assimilation time window (0300 UTC), with the BC observation placed at 0600 and 0900 UTC, respectively. It can be seen that when the BC observation is placed at 0600 and 0900 UTC, positive analysis increments of temperature are generated, with the value of about 0.02 K near the observation location. The mechanism behind the generation of these temperature increments is detailed in Section 3.3.5. This indicates that the temperature of the analysis field will increase due to the assimilation of the BC observation."

Regarding the use of the term "co-correlation," we agree that it is not the most appropriate choice. We have replaced it with "cross-covariance".

39. **Page 19, Line 487: "in fact" should be "in reality" and one can also go on to say "in reality, unlike the single observation experiment, the BC observation is …" to further distinguish the real case from the single observation case.**

Response: According to the reviewer's good instructions, we have revised it as follows:

"In reality, unlike the single observation experiment, the BC observation is …"

40. **Page 20, Lines 507-508: "assimilated all observations within the assimilation time**

window": How frequent is BC observation available for assimilation? I realized that this is actually mentioned in section 3.3.2 that the BC observations are hourly averaged. However, it still didn't say how frequent BC observations are assimilated in the real-case experiments.

Response: Thanks for the insightful comment. In the revised manuscript, we have clarified that in the real-case experiments, the BC observations are assimilated hourly. The revised text now reads as follows:

"…Different from the single observation experiment in Section 5.2, in which the observations are placed at a fixed time, <u>we assimilated all available BC observations with an hourly frequency</u> within the assimilation time window in the full observation experiment…"

41. **Section 4.4: are BC and atmospheric variables minimized together in EXP1 and EXP2 as well? If so, please consider adding a new column in Table 3 to address whether these variables are minimized together or separately. In addition, it might be a good idea to use names that reflects the design of the experiments instead of calling them in numerical order. For example, EXP1 to EXP4 may be renamed to SCDA_BC, SCDA_MET, WCDA_BC+MET, SCDA_BC+MET where SCDA stands for strongly coupled data assimilation while WCDA refers to weakly coupled data assimilation.**

    Response: Thanks for the valuable comment. As we mentioned in our previous response to the general comment, our expression in Section 4.4 in the original manuscript was not sufficiently clear, which may have caused confusion. We apologize for any misunderstanding. After carefully considering the reviewer's comments, we have completely rewritten the original Section 4.4, which is now presented as Section 5.3 in the revised manuscript.

    In the updated version, Table 3 provides a clear description of the four experiments. Additionally, based on the reviewer's suggestion and our revised text, we have renamed the four experiments as DA_BC, DA_MET, DA_MET_then_BC, and DA_MET_BC_simult. We once again thank the reviewer's valuable comments.

42. **Page 21, Lines 517-519: I am not sure if one can really say so without showing results from EXP2.**

    Response: Thanks for pointing this out. Our experimental results show that in the DA_MET experiment, which assimilates operational meteorological observations while excluding BC surface observations, the BC analysis increments are very small. In the revised manuscript, we have added the following clarification:

    "…In the following analysis, we primarily compare the BC analysis increments obtained from DA_BC, DA_MET_then_BC, and DA_MET_BC_simult experiments, <u>noting that the BC analysis increments from the DA_MET experiment are very small (figure omitted)</u> …"

    Additionally, the wording in Lines 517-519 in the original manuscript has been modified as follows:

    "…<u>This indicates that the three BC assimilation strategies have similar assimilation effects on BC, further demonstrating that the assimilation of meteorological observations has a relatively small impact on BC analysis increments</u>…"

43. **Pages 21-22, Lines 539-541: ok, but why? please consider including some interpretation.**

Are BC and atmospheric variables minimized together in EXP1 but separately in EXP3? It doesn't seem quite straightforward and easy to understand, at least to me, why would assimilating only BC observations in a strongly coupled setup leads to similar impact from assimilating both BC and atmospheric observations in a weakly coupled setup? What could be the mechanism that leads to such a consequence?

Response: We sincerely appreciate the reviewer's insightful comments. As we mentioned in our previous response, our expression in Section 4.4 in the original manuscript was not sufficiently clear, which may have caused confusion. We apologize for any misunderstanding. After carefully considering the reviewer's comments, we have completely rewritten the original Section 4.4, which is now presented as Section 5.3 in the revised manuscript.

In the updated version, we provide a clear description of the DA_BC, DA_MET, DA_MET_then_BC, and DA_MET_BC_simult experiments. We specifically clarify that the atmospheric variable analysis increments shown in Figure 10 are solely due to BC assimilation, without contributions from the assimilation of atmospheric observations.

We also explain in detail the reasons behind the similarity and differences in the atmospheric variable analysis increments produced by BC assimilation in the DA_MET_then_BC experiment and the DA_BC experiment. The key points are as follows:

"…This is because, although DA_MET_then_BC first assimilates operational meteorological observations and then BC surface observations, the BC assimilation step only incorporates BC observations, just like in DA_BC. Therefore, the analysis increments of atmospheric variables caused by BC observations in both DA_MET_then_BC and DA_BC are similar. Additionally, the values in each sub-image of the middle panel in Fig. 10 differ slightly from those on the left. These differences are attributed to the distinct basic-state values of the atmospheric variables used in the two experiments. In DA_BC, the basic-state values of the atmospheric variables used in the tangent linear and adjoint processes are derived from the atmospheric background field information without assimilating operational meteorological observations, while in DA_MET_then_BC, the basic-state values are based on the atmospheric analysis field information after assimilating the operational meteorological observations…"

We once again appreciate the reviewer's valuable feedback, which has significantly contributed to improving the clarity and completeness of our study.

44. **Page 22, Lines 553-556: I am not sure if one can make this statement by comparing the *differences* of analysis increments between EXP4 and EXP2 with *actual* analysis increments from EXP1 or EXP3. In addition, I am puzzled while trying to understand how the feedback of BC assimilation on atmospheric variables is reduced by having also assimilated atmospheric observations in a coupled setup without actually seeing the analysis increments in EXP2 and EXP4. Some thought processes and reasonings from the authors are definitely required to be stated.**

Response: We sincerely appreciate the reviewer's insightful comments. We once again apologize for the confusion caused by the unclear description in Section 4.4 of the original manuscript. In the revised manuscript, we have clarified that the atmospheric variable analysis increments shown in Figure 10 are solely due to BC assimilation, without contributions from the assimilation of atmospheric observations. Below, we present the reasons why the analysis increments of atmospheric variables are smaller when both atmospheric and BC observations are assimilated simultaneously:

"…The differences in analysis increments of the four atmospheric variables caused by BC assimilation between DA_MET_BC_simult and DA_BC/DA_MET_then_BC may be due to the fact that information fusion reduces the impact of individual observation. As mentioned above, DA_MET_then_BC is similar to DA_BC in that, in the process of BC assimilation, only BC surface observations are incorporated into the assimilation system. At this stage, the system relies solely on BC observations to correct the initial field. In the absence of atmospheric observations, BC observations play a dominant role, leading to larger analysis increments of atmospheric variables. In contrast, in DA_MET_BC_simult, both operational meteorological observations and BC surface observations are assimilated simultaneously. In this scenario, atmospheric observations provide more comprehensive or reliable information, which may reduce the dominant influence of the BC observations on the analysis increments of atmospheric variables. As a result, a more balanced adjustment of atmospheric variables is achieved in DA_MET_BC_simult…"

We once again appreciate the reviewer's valuable feedback.

45. **Page 22, Lines 556-558: This statement is maybe a little too strong. It sounds like having amplified feedback is not a good thing. Without verifying the analysis with the truth (e.g., re-analysis, or observations that are not assimilated), we do not know if the strongly coupled analysis is actually more accurate than the other ones. Hence, we do not know if amplified feedback is good or not good. Although we'd like to think (or theoretically correct to think) that analysis from a strongly coupled setup is better, we still need some evidence to prove it.**

    Response: We sincerely appreciate the reviewer's thoughtful comments. Indeed, the original wording in Lines 556-558 of Section 4.4 was perhaps too strong, and we apologize for that. In the revised manuscript, we have revised this statement to avoid overly strong assertions. The revised statement is as follows:

    "…As mentioned above, DA_MET_then_BC is similar to DA_BC in that, in the process of BC assimilation, only BC surface observations are incorporated into the assimilation system. At this stage, the system relies solely on BC observations to correct the initial field. In the absence of atmospheric observations, BC observations play a dominant role, leading to larger analysis increments of atmospheric variables. In contrast, in DA_MET_BC_simult, both operational meteorological observations and BC surface observations are assimilated simultaneously. In this scenario, atmospheric observations provide more comprehensive or reliable information, which may reduce the dominant influence of the BC observations on the analysis increments of atmospheric variables. As a result, a more balanced adjustment of atmospheric variables is achieved in DA_MET_BC_simult…"

    We once again appreciate the reviewer's valuable feedback.

46. **Page 23, Line 565: "only 10%": does this mean 10% is not much of an increase? And what is 10% increased computation time relative to? Say, if the microphysics process also takes about 10% computation time, then the readers can have a reference to judge whether 10% is large or small. Without any context, it is just a number.**

    Response: Thanks for the valuable suggestion. We have clarified this point in the revised manuscript as follows:

"the CMA-GFS-AERO simulations increase only about 10% of the computational time of the CMA-GFS simulations (As a reference, the microphysics process accounts for approximately 5% of the total computation time in CMA-GFS simulations) …"

47. **Page 24, Lines 591-592: "three component models" > "three model components"**

    Response: This has been revised as suggested.

48. **Figure 2: I believe the x-axis is missing a base 10 and a minus sign in the power of 10.**

    Response: Thanks for pointing this out. We have revised Figure 2 accordingly and ensured that the x-axis now includes the correct base 10 and minus sign in the power of 10.

49. **Figures S2-S3 and almost all figures: figure captions are rather vague and not very helpful. Both Figs. S2 and S3 present rather complicated ideas and deserve a clearer and informative description.**

    Response: Thanks for the comment. Based on the feedback from Comments #4, #16, and #17, we have removed Figures S2 and S3 in the revised manuscript. Additionally, we have reviewed all figure captions and revised those that were vague to ensure greater clarity and informativeness.

50. **Figure 9: When are these analysis increments valid at? beginning, middle, or the end of the window?**

    Response: Thanks for the comment. We would like to clarify that these analysis increments are valid at the beginning of the window.

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