The authors would like to thank the reviewers for volunteering their time to review this manuscript. Your comments make this manuscript better and better. I have carefully read your valuable suggestions, and the following is my reply.

This manuscript described a process-based budget analysis of the GEFS-Aerosols chemical transport model, including the processes of emissions, reactions and removal. This model budget analysis includes the comparison to the MERRA-2 and GEO4-GOCART, but has few verification with observations, making it hard to evaluate which process has big uncertainties.

Bhattacharjee et al., 2023 (DOI: https://doi.org/10.1175/WAF-D-22-0083.1) evaluated the simulation results of the GEFS-Aerosols model using AOD data derived from satellite retrieval (MODIS and VIIRS), AOD data simulated by other models (MEERA2 and NGAC), and AOD data observed from 50 AERONET stations. The period of evaluation from August 2019 to August 2020 almost coincides with the time period of this study, namely from September 2019 to September 2020. In addition to the regular daily or monthly forecast evaluations of GEFS-Aerosols, three special events were also utilized to evaluate the performance of GEFS-Aerosols. These include dust events in Northwest Africa, agricultural fires in northern India and the August fire complex in northern California.

Zhang et al., 2022 (DOI: https://doi.org/10.5194/gmd-15-5337-2022) evaluated not only the AOD simulated by GEFS-Aerosols from 5 July to 30 November 2019, but also the aerosol concentrations simulated by GEFS-Aerosols during the 22-month ATOM (Atmospheric TOmography Mission) period from 2016 to 2019.

Those sentences has been added to lines 40 to 48 of the manuscript.

The following is the author's response to the detailed comment

Section 2.1 and 2.2. The mass balance equation and associated processes did not mention the model's advection, diffusion, and physical processes. How well has the aerosol mass been conserved in these processes? What's this model's top boundary treatment, and how does the model control the mass leakage through the domain top?

The research object of mass balance equation is the total mass of aerosol in the atmosphere. Any aerosol mass fluxes (e.g. emissions and removals) leaving or entering the system are considered in the mass balance equation. The reaction is also included in the equation as it changes the aerosol species and the mass balance equation is calculated for the aerosol species. Although aerosols are affected by advection, diffusion, and physical processes, these processes are not specifically considered in the equations because they do not cause aerosols to leave/enter the system or change aerosol species. However, these processes do change the concentration of aerosols in the atmosphere, and this effect is ultimately reflected in the two terms "initial and final" in the mass balance equation.

The pressure at the top of the model in GEFS-Aerosols is 200 Pa. As shown in Figures 10 and 11, it is almost impossible for aerosols to be elevated to this level in our simulations,

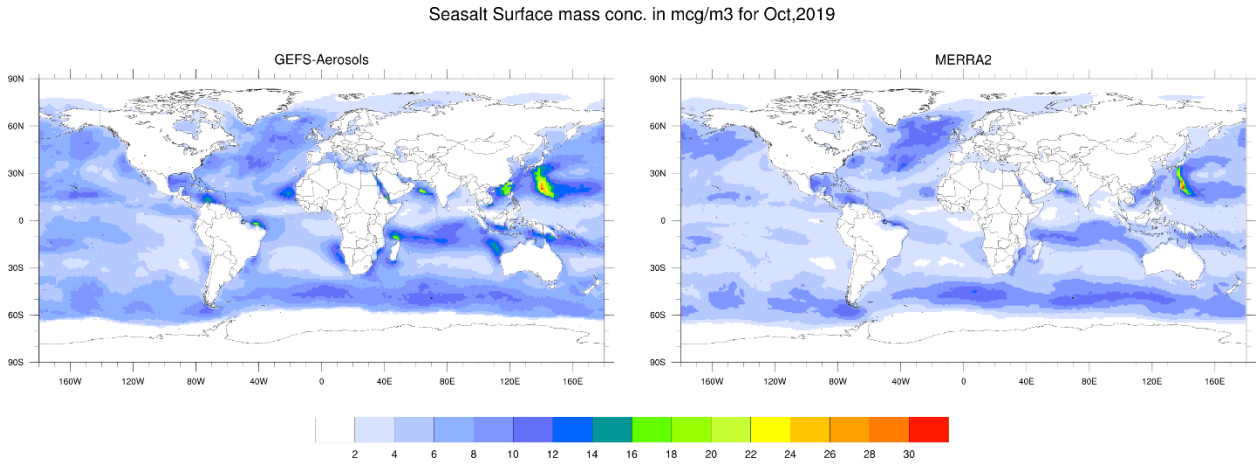
and therefore, at this level, the aerosol concentration in GEFS-Aerosols is the background concentration (1 x 10^{-16} μg/kg).

The gravitational settling of aerosols is usually applied to mass movement from upper layers to the lowest layer of the model, which won't affect the total mass. The removal of aerosol mass from the model's lowest layer to ground surface refers to dry deposition, which should include the gravitational sediment. Does the GEFS-Aerosol's dry deposition scheme exclude the gravitational sedimentation? Please clarify.

The gravity scheme used in GEFS-Aerosol is mentioned in the manuscript lines 76 to 78 " the computation of gravitation settling for dust and sea salt are based on the updated finite-difference scheme in WRF-CHEM (Ukhov et al, 2021)". This scheme not only calculates the gravity settlement from the upper layer of the model to the lower layer of the model, but also calculates the gravity settlement from the bottom layer of the model to the ground. Meanwhile, dry deposition in GEFS-aerosol was calculated by dry deposition velocity based on aerodynamic resistance, sublayer resistance and surface resistance. Therefore, aerosol gravity deposition and aerosol dry deposition are completely separated in GEFS-Aerosols.

Fig 2, and line 115, Are the sea salt AOD calculation method same in GEFS-Aerosol and MERRA-2? Besides AOD comparison, it is better to have mass concentration comparison with observations for sea salt near sea surface. How about the mass flux for other species?

In GEFS-Aerosols, AOD is calculated using a look-up table (LUT) of aerosol optical properties from the NASA GOCART model, so AOD calculations are consistent in GEFS-Aerosols and MERRA2.



The figure above shows a comparison of surface concentrations of sea salt aerosols in GEFS-Aerosols and MERRA2 in October 2019.

Fig 3, 4 and line 117-120. Similar question, does the dry deposition of the lowest model layer exclude the sedimentation? It would better to show the net surface flux of sea salt in GEFS-Aerosol and MERRA-2, to show whether their emission and removal processes are balanced.

It can be seen from Table 2 that the total amount of sea salt emissions and removal processes in GEFS-Aerosols are basically balanced during the one year simulation period.

Fig 6 and corresponding discussion around line 138-145. The model error for dust mass is highly correlated with the dust emission, but these correlations do not exist for BC, OC and sea salt. Any further discussion about the difference among these species, which processes result in the difference?

Indeed, the model errors for BC and OC are also highly correlated with emissions, as indicated in lines 151–152 of the updated manuscript. “The larger model errors for dust, OC and BC coincide with emission outbreaks for dust, OC and BC, which are shown as the emission changes (kg/s -> blue line) in Fig. 6”.

However, the model errors for dust are larger than those for BC, OC and sea salt. The reasons for this were discussed in lines 159 to 164 of the updated manuscript “The assumptions of linearity in aerosol deposition and emissions in the GEFS-Aerosols calculations are the main cause of its model error. This assumption only affects deposition calculations for BC and OC (The daily emissions of BC and OC are constant.), but for dust and sea salt it affects not only deposition but also emissions calculations. At the same time, the wind threshold velocity makes the dust emissions more nonlinear than the source or sink terms of the other aerosol types. Therefore, the model errors for dust and sea salt are higher than those for BC and OC, while the model errors for dust are the highest.”

Fig 10. What is the temporal and spatial extents of these profiles? Please clarify. It is better to separate the profiles over ocean and land, and have a deeper discussion for sea salt and land-source aerosols etc.

Line 282-290. It is more convincing if any direct comparison with observations can be included here, like CALIOP or in-situ measurements.

The data used in Fig. 10 are from September 2019 monthly averages and are global averages. Similar graphs were plotted for other months in this experiment. Since these figures are very similar, they are not shown in this manuscript.

Both Fig. 10 and Fig. 11 represent the simulated vertical distribution of aerosols in GEFS-Aerosols. By comparing Fig. 10 and Fig. 11, we noticed that the different data processing methods when plotting Fig. 10 and Fig. 11 made the GEFS-aerosol vertical profiles in Fig. 10 and Fig. 11 very different. Fig. 11 shows that aerosol concentrations decrease with increasing altitude. However, Fig. 10 shows that at least for some aerosols (e.g. BC, OC, sulfate, and dust) concentrations are higher at higher altitudes. Even so, they are not contradictory. Because Fig. 11 (zonal distribution) is more representative of the vertical distribution of aerosols near the source area (e.g. land), while Fig. 10 is more representative of the vertical distribution of aerosols away from the source area (e.g. over the ocean). Therefore, to evaluate Fig. 11, the HIPPO experiment and CALIOP observations were used, since they measure vertical profiles of aerosols in remote regions.

Added the following sentence into the manuscript: “The decrease in aerosol concentration with increasing altitude shown in Fig. 11 is significantly different from that shown in Fig. 10. Fig. 10 shows that certain aerosols (e.g. BC, OC, sulfate and dust) are more concentrated at higher altitudes. However, Fig. 10 and Fig. 11 are not contradictory. Because Fig. 11 is more representative of the vertical distribution of aerosols near the source (such as over land), while Fig. 10 is more representative of the vertical distribution of aerosols far away from the source (such as over the ocean). Therefore, in the validation of Fig. 10, the HIPPO experiment and CALIOP observations were used because they measured the vertical profile of aerosols in remote areas.”

Table 2 and Section 3.11 It is better to make consistent by changing the units of wet/dry deposition and sediment from percent to Tg/Year, comparable to emission etc.

Three more columns were added to Table 2, indicating the amount of aerosol removed by wet deposition, dry deposition, and deposition (Tg/Y) in GEFS-aerosol, and sentence “and the total removal of aerosols (i.e. the sum of wet, dry and sedimentation) is almost equal to their total emissions (Table. 2).” was added to the text.

Line 350-365. Giving the big discrepancies between the models, is there any observation available to verify aerosol removal process or surface net mass flux?

Measuring aerosol deposition fluxes is extremely challenging (Farmer et al., 2021), so observations are limited. For example, the in-cloud mass scavenging efficiency of BC (Yang et al., 2019), the number of studies in this area is small but the data reported vary widely, making them difficult to use for model evaluation.

In GEFS-Aerosols: “The GOCART dry deposition protocol [Chin et al., 2000] was used for GEFS-Aerosols. Wet deposition in GEFS-Aerosols is the sum of large-scale wet removals and convective scavenging. The large-scale wet removal scheme is from WRF-CHEM (https://github.com/wrf-model/WRF/blob/master/chem/module_wetdep_ls.F), and the convective scavenge is calculated in FV3GFS physics and is based on the simplified Arakawa-Schubert (SAS) scheme [Pan et al, 1994; Zhang et al., 2022b]. The computation of gravitational settling for dust and sea salt are based on the updated finite-difference scheme in WRF-CHEM [Ukhov et al., 2021].”

Farmer, D. K., Boedicker, E. K., & DeBolt, H. M. (2021). Dry deposition of atmospheric aerosols: Approaches, observations, and mechanisms. Annual review of physical chemistry, 72, 375-397.
Yang, Y., Fu, Y., Lin, Q., Jiang, F., Lian, X., Li, L., ... & Sheng, G. (2019). Recent advances in quantifying wet scavenging efficiency of black carbon aerosol. Atmosphere, 10(4), 175.