Response to Reviewer 1:

We would like to thank Reviewer 1 for dedicating time to carefully read our manuscript and provide feedback. We sincerely think their detailed comments have helped us to improve the manuscript. Here it follows a point-by-point response to the reviewer's report (text in black denotes the comments provided, while text in blue denotes our response), associating with the revised manuscript with the track of the changes.

Comments (in black): This study attempted to improve the aerosol simulation in the CESM model by updating the emission schemes for dust and marine aerosols as well as the heterogeneous chemistry of SOA formation. These updates were further supported by evaluating model results against multiple aerosols measurements. Overall, this is an important study and the overall conclusions are reasonable. I would like to recommend its publication after further revisions. In particular, the presentation quality of this manuscript should be improved. Please find my comments in the following:

Response (in blue): We sincerely appreciate Reviewer 1 for the thoughtful suggestions and constructive feedback, which have greatly contributed to the improvement of our paper. In the following responses, we have addressed all the comments and made revisions accordingly, which we believe have led to a significantly improved manuscript.

In Section 2.2, it is strongly suggested to add a Table to list all the model simulations conducted in this study.

Thank you for your constructive suggestion. A new table was added around L283, listing all simulations discussed in the manuscript and their related information, as follows.

Table 1: List of all simulation experiments in this study.

Experiment set	Annotation/Name	Horizontal resolution	Brief descriptions
CYCLE	CYCLE-original	1.9° × 2.5°	2009-2012, CAM6-chem default scheme
	CYCLE-updated	1.9° × 2.5°	2009-2012, updated scheme in this study
Case study of dust events	Case-original	0.9° × 1.25°	1 January 2021 to 1 April 2021, CAM6-chem default dust emission scheme
	Case-updated	0.9° × 1.25°	1 January 2021 to 1 April 2021, updated dust emission scheme
Sensitivity experiments on sea-salt aerosol scheme	SS-Gong	1.9° × 2.5°	2009-2012, *Gong* source function
	SS-Gong+SST	1.9° × 2.5°	2009-2012, *Gong* function together with SST-dependent correction factor
	SS-Gong+RH	1.9° × 2.5°	2009-2012, *Gong* function together with RH-dependent correction factor

Sensitivity experiments on MPOA scheme	MPOA-diatom	1.9˚ × 2.5˚	2009-2012, [Chl *a*] input only from diatom
	MPOA-diazotroph	1.9˚ × 2.5˚	2009-2012, [Chl *a*] input only from diazotroph
	MPOA-small phyto.	1.9˚ × 2.5˚	2009-2012, [Chl *a*] input only from small phytoplankton

Fig.10 compared the simulated and observed sea-salt concentrations, but it looks the model performance is good. This model bias is mainly attributed to the coarse model resolution. Why is model resolution particularly critical for sea-salt simulation? In addition, the updated simulation shows improved correlation but monthly variations of sea-salt aerosols in the two simulations look like very similar. I suggest the authors to check over the calculated correlation.

We appreciate your comments regarding the model bias and the importance of resolution in sea-salt simulations.

1. We agree that model resolution is critical for accurately simulating sea-salt aerosols. The coarse resolution limits the model's ability to capture fine-scale variability in wind speed, which is crucial for simulating sea-salt aerosol generation. This can lead to biases in sea-salt concentration, especially in regions with complex coastal topography or variable oceanic conditions.
   We rephrased the discussion to address these issues as follows:
   L494-499 "*We attribute the bias at these two island stations to the low model resolution. Firstly, the 2-degree resolution used in the model is insufficient to resolve these islands, making it difficult to accurately represent the specific conditions at the stations. Secondly, the coarse resolution results in grid point values that do not accurately reflect the actual conditions, particularly affecting the model's ability to capture fine-scale variability in wind speed. This limitation is critical for simulating sea-salt aerosol generation, as fine-scale wind variations are essential in regions with complex coastal topography or variable oceanic conditions.*"
2. Regarding the similarity in monthly variations between the two simulations, we acknowledge that while the correlation improved, the overall pattern remained consistent between the original and updated schemes. This is likely because the primary source functions and meteorological inputs, which drive the monthly variability, are similar in both simulations. We have re-examined the calculated correlation (R) to ensure the accuracy.

As shown in Fig.15, the effects of added SOA formation pathways are notable over biogenic and biomass burning-affected region. As such, I suggest to show some seasonal variations of these effects.

Thank you for your insightful suggestion. We added some descriptions in L609-614 and a figure in SI (Figure S9) to show the seasonal variations of these effects.
"*In addition, we find that the effect of dicarbonyls on SOA formation shows significant seasonal*

variation, with higher contributions in boreal summer (JJA) and winter (DJF) and relatively lower contributions in spring (MAM) and autumn (SON). Regionally, high values in summer are mainly observed in Southeast Asia, North America, and the Amazon, while in winter, they are concentrated in Central Africa and South Asia (Fig. S9a). Biogenic emissions of isoprene, the primary precursor of dicarbonyl compounds (Fu et al., 2008; Kelly et al., 2018), are the main drivers of these spatiotemporal variations (Fig. S9b)."

In Abstract and Conclusions Sections, I strongly suggest the authors to add some quantitative conclusions that show the effects of update schemes on aerosol simulations.

Thank you for your constructive suggestion. In the revised manuscript, we incorporated specific numerical results to provide a clearer understanding of their significance.

Abstract. "*Aerosols constitute important substance components of the Earth's atmosphere and have a profound influence on climate dynamics, radiative properties, and biogeochemical processes. Here we introduce updated emission schemes for dust, sea-salt, and marine primary organic aerosols (MPOA), as well as augment secondary organic aerosol (SOA) formation pathways within the Community Earth System Model (CESM; version 2.1.3). The modified dust emission scheme shifts the original hotspot-like dust emission to a more continuous distribution, improving the dust aerosol optical depth (DAOD) simulations at stations in North Africa and Central Asia. This update also reduces dust residence time from 4.1 days to 1.6 days, enhancing concentration simulations downwind of dust source regions. For sea-salt emissions, we incorporate an updated sea surface temperature (SST) modulation and introduce a relative-humidity-dependent correction factor for sea-salt particle size with SST having a significantly larger impact on sea-salt emissions (16.1%) compared to the minor effect of humidity (-0.3%). We then extend to incorporate emissions of marine primary organic aerosols (MPOA) as externally mixed with sea-salt aerosols, coupled offline with ocean component Parallel Ocean Program (POP2). The results underscore the substantial influence of phytoplankton diversity on MPOA emissions, with 148% variability simulated among different phytoplankton types, highlighting the role of biological variability in aerosol modeling. Furthermore, we refine the model's chemical mechanisms by including the irreversible aqueous uptake of dicarbonyl compounds as a new pathway for SOA formation, contributing an additional 37% to surface SOA concentrations. These improvements enrich the capability of the CESM to use intricate linkage between different components of the Earth system, thereby enabling a more comprehensive description of natural aerosol emissions, chemical processes, and their impacts.*"

4 Summary and conclusion

"*This study sets out to develop updated emission schemes for natural aerosol species based on the CoAerM, including dust, sea-salt, and MPOA, and SOA formation, including an irreversible aqueous uptake of dicarbonyls, in the CESM2. For dust emissions, the modified scheme confines dust deflation to erodible areas based on land use distribution instead of the original geomorphology-based hotspot-like source function, and integrates reduction factors for vegetation effects. Roughness length and soil texture from the land component, CLM5, is also incorporated to update threshold friction velocity correction factors. The updated*

scheme yields a more continuous distribution of dust emission areas, and complements the emissions in North America and the sub-Arctic. Notably, DAOD simulations at stations in Central Asia (Karachi) and North Africa (Tamanrasset_INM) show more consistent alignment with observations in the updated scheme. Also, the updated scheme acts to shorten the residence time of dust aerosols from 4.1 days to 1.6 days, resulting in notable changes in simulated dust burden and associated DAOD simulations, particularly in downwind areas of the dust source region. The simulation of dust aerosol concentrations during dust events is improved by the updated scheme in the downwind region of dust propagation. The sea-salt emission scheme is modified through updating the dependence of source function on SST and introducing a relative-humidity-dependent correction factor for sea-salt particle size. These modifications align emissions more intuitively with oceanic conditions and sea-salt production mechanisms. The modulation of sea-salt emission by SST is more pronounced in the simulations of the updated scheme, resulting in an increase in sea-salt emission in the tropical and subtropical oceans and a decrease in the Southern Ocean. The RH correction factor exerts an enhancing effect across the globe, but the effect is very mild, resulting about 0.3% decrease in sea-salt emissions.

Moreover, we extend CESM's capabilities to capture the link between marine biology and atmospheric chemistry by including the MPOA emission scheme. Coupled offline with ocean component POP2, the representation of phytoplankton chlorophyll distribution by the ocean biogeochemistry module, MARBL, plays a crucial part in modelling MPOA emissions. In our simulation, the total global mass of MPOA emitted during 2010-2012 is 8.5 Tg per year. Our simulations reproduce the seasonal cycle observed at the North Atlantic station (Mace Head). However, the bias in the simulation of the peak month at the Southern Ocean station (Amsterdam Island) may be related to the model's simulation of the dominance of small phytoplankton in this region. We further compare the spatial variability of different phytoplankton species on MPOA emission simulations, highlighting the significance of biological diversity in shaping aerosol emissions, with a 148% variability simulated among different phytoplankton types. For the formation of SOA, we consider the irreversible aqueous uptake of dicarbonyl compounds (glyoxal and methylglyoxal) in the chemical mechanism. The results show that this pathway makes an important contribution to the surface SOA concentrations (an additional 37% surface SOA concentrations), especially during severe haze events. The accurate simulation of SOA needs further research into incorporating additional processes and optimizing model parameters. Collectively, these modifications make the CESM a comprehensive tool for elucidating the complexities of aerosol emissions and transformation from different spheres in the Earth system, such as the land and ocean, thus facilitating the potential for improved evaluation of their impacts on climate processes and feedback."

The table caption should be moved to the top of each table.

Corrected throughout the manuscript.

I think the "CAM6-Chem" should be changed to "CAM6-chem" throughout the text.

We agree and have thoroughly re-examined the use of the terms throughout the revised manuscript.

L263: The reference for MERRA2 reanalysis data looks not correct.

We reviewed the citation requirements for this dataset, and the corrections are as follows:
(NCAR/UCAR, 2018)
Atmospheric Chemistry Observations Modeling/National Center For Atmospheric Research/University Corporation For Atmospheric Research and Climate And Global Dynamics Division/National Center For Atmospheric Research/University Corporation For Atmospheric Research: MERRA2 Global Atmosphere Forcing Data [data set], https://doi.org/10.5065/XVAQ-2X07, 2018.

L267: The "CYCLE" simulation was using CAM5 not CAM6?

We apologize for the misunderstanding caused by the typo here. The correct term is CAM6-chem, and this has now been updated in the text.

L302-303: I suggest to move the introduction of model evaluation metrics after Section 2.3.

We reorganized Section 2.3 to place the introduction of model evaluation metrics at the end of this section:
L327-329 "*In the following discussion, the evaluation metrics used are the Kendall's correlation coefficient (R) and root mean square error (RMSE). Kendall's correlation, which does not assume a specific data distribution, is used to assess the statistical dependence between observed and simulated values. RMSE measures the average error between observation and simulated results.*"

L316: Why were these two measurement sites considered here?

Thank you for your question regarding the selection of the two measurement sites. Mace Head and Amsterdam Island were chosen due to their geographical locations and the availability of long-term observational data. Mace Head represents a Northern Hemisphere mid-latitude site, while Amsterdam Island provides a contrasting Southern Hemisphere marine environment. These sites offer valuable insights into the performance of our model across different hemispheres and oceanic conditions. Additionally, these sites are commonly used in previous studies for comparing MPOA simulations, making them suitable for consistent and meaningful evaluation of our model.
We clarified this rationale in the revised manuscript as follows:
Starting from L564 "*We also evaluate the model simulation of MPOA concentrations using measurements from two representative sites. The first site, Mace Head (53.33˚N, 9.90˚W), located near biologically productive waters in the North Atlantic Ocean……Another observation site is Amsterdam Island (37.80˚S, 77.57˚E), which is subject to windy and biologically active currents in the Southern Ocean……*"

L461: Any updated model results from CMIP6?

Thank you for pointing this out. We agree that referencing the most recent CMIP6 data would provide additional context and situate our findings within the latest climate modeling framework. We included a comparison with the updated CMIP6 model results in the revised manuscript as follows:

L463-466 "*Nevertheless, our results fall in the mid-range of values estimated from the historical CMIP5 simulations compiled in IPCC AR5 (1400-6800 Tg/year) (Intergovernmental Panel on Climate Change (IPCC), 2014), and are also consistent with the broader range observed in CMIP6 simulations (2624–64939 Tg/year) (Thornhill et al., 2021).*"

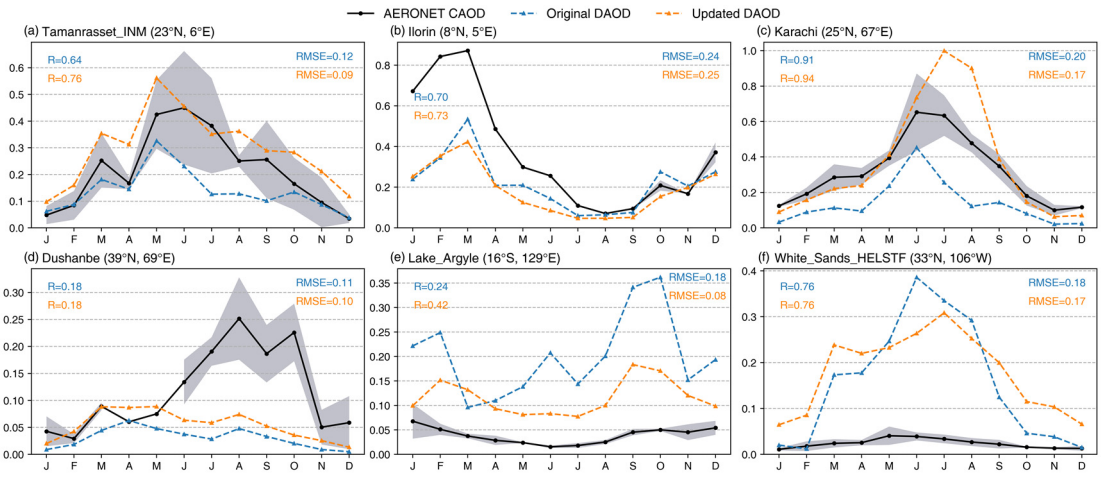
L610: The authors only show the changes in global annual mean PM2.5 but attempt to link it with haze episodes. Any further evidence to support this argument?

We clarified this by modifying the sentence in L630-636 as:

"*Previous studies have shown that during heavy haze episodes, organic aerosols can account for up to half of the PM mass, with a significant contribution from SOA (Huang et al., 2014, Zhao et al., 2019). Figure 16 shows the simulated global monthly surface mean concentrations of SOA and PM2.5 during 2010-2012. The model suggests that the irreversible aqueous uptake rate of dicarbonyls increases notably (solid black line) when heavy haze events occur, resulting in a strong increase in SOA concentrations. The results indicate that the aqueous pathway through dicarbonyls can improve the underestimation of observed SOA concentrations during severe haze episodes (Li et al., 2019; Li et al., 2021).*"

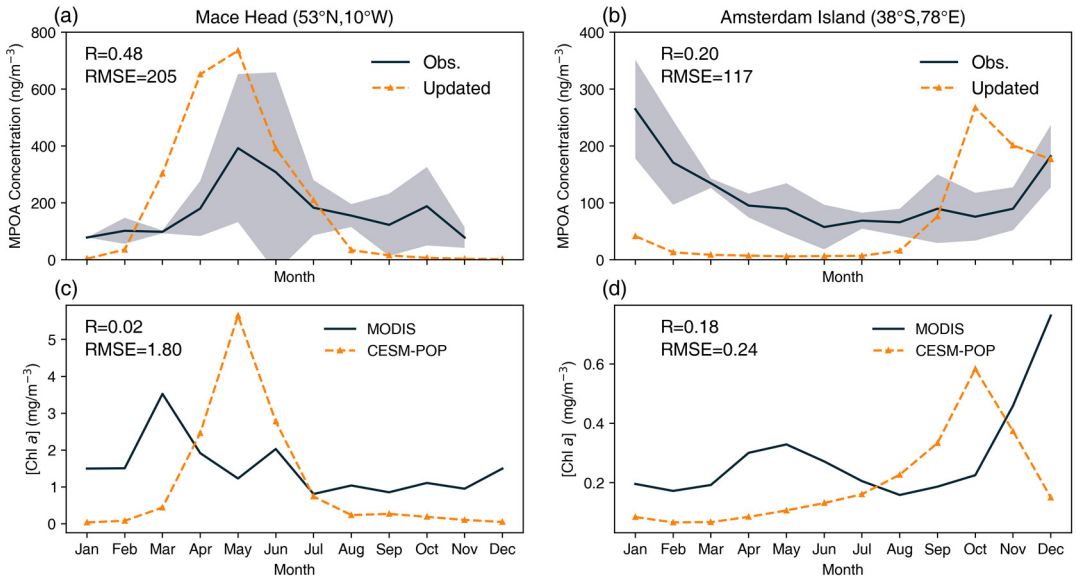
In Fig.6, I suggest to add the location information (e.g., Latitude and longitude) of each AERONET site on the plots.

Thank you for your suggestion. We incorporated this information in the revised manuscript to enhance the clarity and utility of the figure.



In Fig.13, to be consistent with other Figures, I suggest to also add the RMSE metric in the plot.

Thank you for your suggestion. We added the RMSE metric in Fig. 13 (also the site location information) to enhance consistency across the figures and allow for easier interpretation of the results.



Additional changes:

- Minor wording adjustments and corrections throughout the manuscript.
- Added annotation to the right side of each panel in Figure 4, 9, and 15 to provide clearer context.
- Standardized the formatting of PM2.5 in the text to display as $PM_{2.5}$ in subscript.

We hope that we have adequately addressed all the suggestions raised by Reviewer 1, and appreciate their constructive feedback