Comment on acp-2021-409
Anonymous Referee #1

Referee comment on "Quantifying the structural uncertainty of the aerosol mixing state representation in a modal model" by Zhonghua Zheng et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-409-RC2, 2021

The authors verified the global distribution of aerosol mixing state represented by modal models, focusing on comparing the calculations from ML model with MAM4. The author concluded the current model simulations on aerosol mixing state had large and zonally structured errors, and ML model trained on high-detail particle resolved simulations were more representative for realistic aerosols. The technical analysis is of reasonably high quality. The interpretation and discussion could benefit from some stronger quantitative analysis. The content is suitable for publication within the scope of ACP, while some revisions are required. Please see detailed comments below.

General Comments:

Although ML model trained on high-detail particle resolved simulations is better than MAM4 for simulating aerosol mixing state, it still has some uncertainties which have not mentioned in this work. Taking the mixing of optically absorbing and non-absorbing species ($\chi_o$) for example, the ML simulations in this work are in the range of 50-90%, significantly higher than the realistic aerosols due to greatly underestimate of BC-free particles in some regions. Earlier work (Figure 8 in Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-222, 2017) reported that 85%-90% of particles in the accumulation mode were BC-free particles, and internally mixed BC only accounted for 5-10% during summertime in North China Plain. The significantly underestimate of BC-free particles in ML simulations in this work is most likely attributed to irrespective of the processes of new particle formation and growth from Aitken mode to accumulation mode, which are hardly associated with particles (i.e., BC and POM) from primary emission. In term of the mixing of primary carbonaceous and non-primary carbonaceous species ($\chi_c$), the processes of new particle formation and growth will cause a similar uncertainty like the $\chi_o$. Therefore, in the regions where particles are dominated by new particle formation and growth processes, the methods used in the ML simulations in this work should be improved. The author should clarify this uncertainty as discussed above and give some suggests how to improve their ML simulations.

Specific comments:

Page 1/Line 7: The abbreviation can not be used here. Please give the full name of MAM 4.
Page 4/Line 88: Why the authors ran the model for the year 2011 rather than more recent years?

Page 5/Figure 1: In terms of the size distribution of the four modes (i.e., left panel), please show the diameter values of x-axis. If so, the readers can clearly know the size ranges of the Aitken, accumulation, primary carbon and coarse modes.

Page 6/Line 133: not "rather then", here should be "rather than".

Page 6/Line 140: using a terminology as "mass absorption cross section" rather than "a mass absorption coefficient".

Page 7/Line 164 and Line 176: For the input parameters of primary emissions of gas phase, the authors consider the NO$_2$. Why not considering the nitrate in the aerosol species?

Page 7/Line 182: Please claim how to calculate the errors, namely 5% for $\chi_o$ and 8% for $\chi_c$ and $\chi_h$. What are the sources of the error? Why so small errors for model simulations?

Page 11/Line 248-250: The authors claim that their simulations of ML model can not well represent the mixing state of sea salt. This will cause how much errors in the ocean regions?

Page 13/Line 279-284: The authors state that the threshold is set to a relatively large value in MAM4 and their results are a reflection of this fact. Could the authors provide a reasonable value or range of the threshold based on their results?

Page 15/Line 326-327: Here do not need the full name of $\chi_o$, $\chi_c$ and $\chi_h$.

Page 17-18/Figure A1 and Figure A2: The authors use a1, a2 and a4 represent different modes? Why is there no a3?