

We thank the reviewers for their careful reviews and helpful comments. The manuscript has been revised accordingly and our point-by-point responses are provided below. (Reviewers' comments are in italic and the responses in standard font).

Reviewer #1

The authors present a 4-mode modal aerosol model (MAM4) by introducing a primary carbon mode to the existing 3-mode version of MAM (MAM3) in the Community Atmosphere Model version 5 (CAM5). They further design two sets of sensitivity experiments, one to change the aging properties of primary carbon and another to change model resolution, to investigate the potential improvement of atmospheric black carbon simulation. The paper is well written. I recommend publishing the paper on ACP after the authors make some minor modifications.

General Remarks:

My only concern is the performance of MAM4 relative to MAM3. The authors evaluate BC results simulated from MAM3 and two sets of sensitivity experiments of MAM4 using various aircraft measurements. With the exception of high latitudes of the Northern Hemisphere, the performance of MAM4 BC simulation over global broad coverage is deteriorated, overestimating BC compared to measurements. Over the North Pole region, MAM4 BC is still underestimated and the authors suggest further improvements on in-cloud scavenging and vertical transport in convective cloud and on emissions. These actions, while they can potentially improve BC over North Pole, are very likely to further downgrade BC simulation outside North Pole regions.

Reply: We agree with the reviewer that the primary goal of MAM4 is to improve the BC/POM simulation in remote places away from the source regions by treating the aging process of BC/POM that is neglected in MAM3. However, because the current model is calibrated for MAM3, running the model with MAM4 results in the overestimation of BC concentrations in the free troposphere in the regions evaluated in this study (e.g., remote Pacific during the HIPPO campaign, and tropics, NH subtropics and NH mid-latitudes during the AVE-Houston, CR-AVE, and TC4 campaigns).

We suggest a potential further improvement of in-cloud scavenging and convective transport of aerosols in the manuscript. As shown in H. Wang et al. (2013), with a unified treatment of in-cloud scavenging and vertical transport in convective clouds, the overestimation of BC in the free troposphere in the remote regions (e.g., over the Pacific) is significantly reduced, due to the inclusion of secondary activation and thus more efficient scavenging of aerosols in convective clouds. Meanwhile, H. Wang et al. (2013) found that the liquid cloud fraction in the NH mid- and high latitudes is too high, which leads to too efficient scavenging of BC during its transport from the NH mid-latitudes. By modifying the treatment of liquid cloud fraction in cloud scavenging parameterization for the NH high latitudes, H. Wang et al. (2013) gave a much better simulation of near-surface BC and other aerosol species in the Arctic than simulation from the standard CAM5.

We also suggest a further improvement on aerosol emissions. As pointed out by Stohl et al. (2013), BC emissions in the NH high latitudes are underestimated and gas flaring emissions are omitted in the IPCC AR5 emission inventory used in CAM5 simulations. We anticipate that the low BC bias in the Arctic can be reduced by improving the emission inventory. We note that improvements in emissions in the NH high latitudes (in and near the Arctic) would not necessarily downgrade the simulation outside North Pole regions.

A more recent improved model treatment in the resuspension of aerosols from evaporated raindrops, releasing aerosol particles to the coarse mode instead of their originating mode upon complete evaporation of raindrops (Easter et al., 2015, personal communication), has shown a significant impact on the vertical distribution of aerosols, including large reductions in mid- and upper tropospheric BC and POM.

The manuscript has been revised in Section 5 to include the discussion above.

Specific comments:

1. Page 8342 lines 22-23: Overestimating BC over Pacific region is a common problem for many global aerosol models. Changing emission based on various available emission inventories cannot solve the problem.

Reply: As discussed in the manuscript, the overestimation of BC over the Pacific can be mitigated by the improved treatment of in-cloud scavenging and vertical transport in convective clouds, as shown in H. Wang et al. (2013). Other model improvements in cloud processing of aerosols (e.g., Easter et al., 2015, personal communication) would also help reduce the high bias. The low BC bias in the Arctic is much reduced with MAM4, although BC concentrations are still lower than observations. Stohl et al. (2013) pointed out that BC emissions in the NH high latitudes are underestimated in the AR5 emission inventory. We anticipate that the low BC bias in the Arctic can also be reduced by improving the BC emissions.

2. Page 8343 line 22: Add Bian et al., 2013 alone with Wang et al., 2013.

Reply: Done.

3. Page 8343 line 23: Add Jiao et al., 2014 after “dry and wet deposition”.

Reply: Done.

4. Page 8344 line 1: Add “and II” and “Phase I”.

Reply: Done.

5. Page 8344 line 1: Add “Samset et al. 2014” after “Schwarz et al., 2010”.

Reply: Done.

6. Page 8344 line 11: Add “nitrate” after “sulfate”.

Reply: Currently MAM does not treat nitrate. To avoid misunderstanding, we prefer to remain the original wording, “(e.g., sulfate)”.

7. Page 8344 lines 15-16: Does “its” refer to BC’s? If yes, why is “BC’s” absorption of sunlight enhanced significantly since soluble species, typically sulfate and nitrate, have less absorption than BC’s?

Reply: Yes, “its” refers to BC’s. We have explained this more clearly in terms of the lensing effect for the shell-core treatment (Jacobson 2001, 2003), and in terms of increasing the cross section of the absorbing material for the volume mixing treatment (Adachi et al., 2010). We’ve also added a paragraph in section 2.3 on the volume mixing treatment used in MAM4.

8. Page 8345 lines 19-24: *If these are the reasons for the underestimation of BC at high latitude of Northern Hemisphere, then how do the authors explain the overestimation of BC over other regions?*

Reply: In the manuscript we describe the CAM5 (with MAM3) low bias for near-surface BC concentrations in the Arctic, and review previous studies that attribute the bias to emissions, wet scavenging parameterization, and model resolution. The overestimation of BC occurs in the upper troposphere in other regions (e.g., remote Pacific), which is another important model bias. We believe that this high BC bias in the upper troposphere is largely due to the treatment of vertical transport and wet removal in convective clouds. A revised treatment of these processes in H. Wang et al. (2013), which included secondary activation in convective updrafts, noticeably reduced this upper-troposphere high bias with MAM3. This revised treatment was not used in our simulations because it is not included in the standard CAM5 code yet.

9. Page 8346 lines 10-12: *How does aerosol affect convective cloud?*

Reply: In CAM5, aerosol does not affect the microphysical processes of convective clouds. We have made it clear in the revised manuscript.

10. Page 8348 lines 8-11: *Do the authors use different hygroscopicity for fossil fuel POM and biomass burning or use the same value for both?*

Reply: The hygroscopicity of POM from fossil fuel and biomass burning sources is set to be the same in this study. We have made it clear in the revised manuscript.

11. Page 8349 lines 13-15, Page 8350 lines 3-4, and Figures 6-14: *How large is the inter-annual variation of BC over the comparison regions? The current approach of the*

comparison implies that the inter-annual change of BC is very small.

Reply: As described in Section 3, our set 1 experiments were conducted using the present-day (i.e., year 2000) climate forcing conditions for 11 years with the last 10-year results used for analysis. It is not an AMIP-type configuration so a specific model year does not represent any particular calendar year. The 10-year average is viewed as an ensemble mean, representing the model's present-day climatology. Ideally the model climatology should be evaluated against the corresponding observational climatology. However, the field campaigns were conducted in a particular time period and such a comparison is not feasible. Our purpose is to provide the model climatology from the sensitivity study and use the aircraft measurements as a reference, and acknowledge that the observation represents a particular year while the model results are climatology. Therefore, the inter-annual variation of BC due to meteorology and/or forcings (e.g., aerosol emissions) cannot be quantified from such experiments. Other than these factors, the inter-annual variation of modeled BC due to natural variability (i.e., BC difference among different years of 10-year climatology) is quantified to be very small, as indicated by the standard deviations of BC/POM budget terms about the 10-year mean in Tables 2 and 3 for the set 1 experiments (added in the revised manuscript).

By contrast, our set 2 experiments are nudged toward the year of 2009 meteorology. The differences in BC from MAM4L8 of set 1 and MAM4R1 of set 2 experiments are due to differences in their meteorology (winds, clouds, and precipitation), which include the differences between a 10 year average (or ensemble) and a single year, as well as the differences between the free-running model's meteorology and the nudged model's meteorology (which is very close to the reanalysis).

We have revised the manuscript to clarify the model-to-observation comparison strategy.

12. Page 8354 line 19: Add Bian et al., 2013 after Ma et al., 2013.

Reply: Done.

13. Figure 6: Why is MAM3 result not shown on the figure?

Reply: The MAM3 result is already shown in Tilmes et al. (2015). We have added a sentence to direct readers to Tilmes et al. (2015) for MAM3 result.

14. Figures 7-12: How are the model results sampled spatially and temporally when they are compared with observations?

Reply: The model results are sampled in the same way as Liu et al. (2012): Simulated profiles are averaged over the locations on the map and the indicated month of respective field campaigns (in Figures 7-12). This sentence has been added to the figure captions.

Technique corrections:

Page 8344 lines 4-5: Change “compared” to “comparable” and delete “the models tend to be in better agreement”.

Reply: Done.