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**Geoscientific
Model Development
Discussions**

Interactive comment on "Sensitivity of remote aerosol distributions to representation of cloud-aerosol interactions in a global climate model" *by* H. Wang et al.

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We thank the two anonymous referees for their valuable comments to the manuscript and their constructive suggestions for improving the presentation quality. Below, we explain how the comments and suggestions are addressed and make note of the changes we have made to the manuscript.

Anonymous Referee #1:

This paper uses observations of BC in combination with a sophisticated climate model (PNNL-MMF) capable of resolving sub-grid cloud processes to diagnose the sensitivity of remote BC concentrations in the tropical high troposphere, and polar regions to

changes in bulk cloud-processing parameters in the global atmospheric model CAM. They find that inclusion of a new unified treatment of vertical aerosol transport and convection (increasing the scavenging rate of BC in the tropical troposphere) improves the models agreement with high altitude observations of BC. While, at the poles, BC concentrations increased 10-fold in winter due to a combination of reductions in the frequency of liquid-phase cloud scavenging in the mid-to-high latitudes and an increase in the BC ageing lifetime. Inclusion of these 'improved' parameters significantly improved the agreement between the PNNL-MMF climate model and CAM. However, winter-spring concentrations in the Arctic are still significantly underestimated in both models. This study is extremely broad and in some places quite difficult to understand although the conclusions are consistent with previous work in the field. Below are some suggestions to improve the clarity of the paper.

1. Line 17 page 333, over-predict should be over-predicts

AR: Corrected.

2. Line 11 page 337, BC and POM is emitted into the accumulation mode in MAM3 but a primary carbon mode in MAM7. How large is this primary mode and is the biomass and anthropogenic component separated? Are BC/POM particles transferred directly to the accumulation mode after ageing or to the soluble Aitken? In general the paragraph beginning at line 5 needs more detail. For reference it would be useful to have a typical size-range included for each mode i.e. Aitken (xnm-ynm). If primary BC is emitted into an Aitken primary mode how does the emission size impact the studies result (i.e. does the increase in BC stem from ageing or from a reduction in the average size of carbon particles?)?

AR: The BC and POM particles have the same size distribution upon emission in MAM3 and MAM7 (σg=1.8; number mode diameter of 80nm). Thus the emission size does not directly cause the differences between MAM3 and MAM7. However, in MAM3, freshly emitted BC and POM particles are immediately internally mixed with other aerosol hy-

groscopic species (sulphate, SOA, sea salt) present in the accumulation mode. In MAM7, BC and POM are emitted into a primary carbon mode, which has a low hygroscopicity. If we define size range for particle diameters as the 10th and 90th percentiles of the global annual average number size distribution (Liu et al., 2012), the Aitken-model particles are in the range of 15-53 nm and accumulation-mode particles are in the range of 58-270 nm, and the MAM7 primary-carbon-mode particles are in the range of 39-130 nm. The primary-carbon-mode particles subsequently age by condensation of soluble material (sulphate, ammonium, SOA) and coagulation with other soluble particles, and are gradually and directly transferred to the accumulation mode where they mix with other species. We have added more information about aerosol parameterizations to the revised manuscript as suggested (e.g., the prescribed geometric standard deviation, σg, size range for each of the log-normally distributed modes for MAM3 and MAM7, BC/POM particle size upon emission, etc.).

Biomass burning and anthropogenic components of BC and POM are not separated in the MAM configurations used in this study, but they have been separated in some other MAM studies (e.g., Ghan et al., 2012).

3. Line 24 page 339, What do the authors mean by surface wet-deposition flux? Is this the mass deposited to the surface per day or the BC mass in the model surface layer deposited per day? Does this flux include a mixture of impaction/in-cloud scavenging? Or in-cloud (nucleation scavenged) only. When discussing scavenging processes please indicate the type each time.

AR: By surface wet-deposition flux, we meant the mass deposited to the surface per unit area per day. This flux does include contributions from in-cloud scavenging and below-cloud scavenging (impaction and Brownian diffusion). Amongst the several wet removal processes that occur in and below convective and stratiform clouds, the most efficient one for submicron aerosol (such as BC and POM particles) is nucleation scavenging in liquid clouds. We now make this clear at various places in the revised manuscript.

4. Page 339-340, I am assuming that the wet deposition flux is for the total column as Fw,ta has the same subscript as Bta (although it is unclear why this subscript is present). Unless the subscript is explained I would suggest rewriting the formula as: Rw = Fw/B. In general this description is too vague, all terms need to be clarified.

AR: The subscript ta (standing for total aerosol), was used to distinguish from ca (standing for cloud-borne aerosol) in Eqs. 1-3. We agree with the referee that the two-layer subscript, “w, ta”, looks quite redundant. We now take the suggestion and simplify the denotation of terms in all three equations by removing the subscript ta, and clarify them more in the revised manuscript.

5. Figure 1 (Figure 2): Fractions should be shown on a linear scale. A plot of LWC would also be useful here.

AR: Fractions in Figs. 1e,f and 2e,f are now shown on a linear scale.

Including plots of LWC in Figs.1 and 2 was an excellent suggestion. They have been added and connected to relevant discussion in the revised manuscript. We also had a plot of LWP in Fig. 9 and some discussion on this.

6. Equations 1 - 3 pages 339-340, The methodology described here is extremely opaque and requires clarification. Is the first term of this equation (Fw,ta/Bca) calculated as the autoconversion rate of the cloud-water or as the modeled deposition flux over the calculated burden? It is my understanding that the aerosol burden in models is calculated from model output after the deposition flux has been subtracted. If this is the case than the first-order wet removal will be biased high. Please explain where these terms come from.

AR: We have substantially modified section 2.2 to more clearly explain the methodology used in Eqs. (1-3) and Figs. 1 and 2. As part of this, we explain how the individual terms are calculated (“where they come from”).

Regarding Eq. (1), B is obtained from monthly-average mixing ratios at each model

grid-point, which are integrated vertically then further averaged over multiple simulation months (e.g., DJF) and years. Fw is obtained from monthly-average wet-deposition fluxes for each vertical column, and then further averaged in the same way as for B. (Note however that wet removal and precipitation-borne aerosol sedimentation fluxes are calculated at each level in the model.) Rw, which is derived from the averaged B and Fw values using Eq. (1), represents the inverse time-scale for wet-removal of the column BC burden. Similarly, the three terms on the right-hand-side of Eq. (2) are calculated from time-averaged quantities (Fw , Bc, B, fct) so that are equal at each model grid-point (or grid-column). These terms could be calculated for each time-step, and then averaged; this would give somewhat different results numerically. However, since the purpose of these diagnostic quantities is to provide insight into differences between models and simulations, and inform us of sources of uncertainty, these computational nuances are of secondary importance.

Regarding the ordering of calculations and the potential high bias in the first-order wet removal rate, the referee is correct here in that some high bias is likely present. The purpose of our methodology and analysis is to perform a quantitative comparison among simulations of factors that affect BC wet removal (and thus transport to high latitudes) in the model. Biases resulting from how the various terms are calculated will affect the accuracy of the comparison, but we feel that the methodology still provides useful insights.

7. Line 4 page 343, please explain the meaning of wet-removal adjustment factors.

AR: These adjustment factors (which have magnitudes $\leq$ 1) are tunable parameters applied to the calculation of stratiform/convective in-cloud and below-cloud scavenging rates to account for various uncertainties in the model predicted aerosol activation, below-cloud scavenging efficiency, cloud fraction, cloud water, and precipitation. In one of our sensitivity tests, CONV_SF, the stratiform in-cloud wet removal adjustment factor was reduced from 1.0 to 0.6 to account for the too rapid cloud water to rain conversion in CAM5.

8. Table 1, This table is not useful it would be better as a grid with the model simulations on the left and the new parameters at the top with symbols (i.e. tick marks) showing which model improvements are in which simulation.

AR: This constructive suggestion has been taken, and we revised the Table 1.

Anonymous Referee #2:

The paper presents some new developments implemented in the global atmospheric model CAM with the objectives to improve the representation of aerosols in regions that are usually poorly represented in models, including the upper troposphere and the Arctic. The CAM model is bench-marked against a model version that includes a more detailed representation of cloud-aerosol interactions through a sub-grid scheme and against observations. The inclusion of a new unified treatment of vertical aerosol transport and convection results in an improvement of the model comparison against observations.

The paper is overall clear and well written and is very well suitable for GMD as it describes new model developments that could serve as a useful basis for users of the CAM model and of other models. I do not have any major comment about the manuscript and only small clarifications are suggested below.

Specific comments:

- P342, lines 10-15: Provide some key aspects of the new unified treatment as explained in S1. What are the main new features of this “improved unified treatment”? (Include for example indications such as those provided in lines 58-62 of S1).

AR: This is an excellent suggestion. We have now added the key aspects of the new unified treatment to the main text (section 2.3).

- P342, lines 22-30: It is not clear whether a new scheme was applied in this CONV_FD simulation, or whether different parameters were applied. In any case, what are these parameters?

AR: The “freeze-dry” scheme is an existing parameterization in the model, but it is not used by default. The comparison between default CAM5 and MMF suggests that liquid cloud fraction in super-cooled environment (usually the specific humidity is small) is over-predicted in CAM5. This was also found by Vavrus and Waliser (2008) who developed the “freeze-dry” scheme to correct the over-prediction by an empirical factor of max[0.15, min(1,qv/qv0)]. The “freeze-dry” scheme uses a threshold specific humidity qv0=0.003 kg kg-1. We doubled this threshold to further reduce liquid cloud fraction in a broader area in CAM5, getting closer to the MMF liquid cloud fraction (see Fig. 2f). We also noted in the manuscript that alternate approaches for achieving lower liquid-cloud fractions at cold temperatures may be more desirable.

- P343: line 5: What are these adjustment factors? How are they “adjust”?

AR: Please see the response to the same comment (#7) from referee #1.

- P366, table 1: Please clarify which simulations use the m7 or m3 configuration.

AR: MAM3 is the default model setting, so “m3” was omitted in the simulation names. We have revised Table 1 to include the aerosol configuration and also made this clear in the text.

- Section 3.4: The discussion about the differences between the M3 and the M7 configurations are quite interesting, as it touches upon the level of complexity that is needed in climate models if one wants to represent aerosol processes and their impact on climate. I understand that some of these issues are discussed in other papers but I still think it would be useful to summarize more clearly in this paper the “pros and cons” of using the MAM7 versus MAM3 to represent aerosols burdens in the upper troposphere and the remote regions. Is the slower aging assumption the main difference?

AR: We agree that higher level of complexity and sophistication in the aerosol treatment doesn’t mean better representation or performance in every aspect. For example, the 7-mode aerosol treatment reduces the high-latitude BC low biases, but worsens the

over-prediction of upper-tropospheric BC. We have added more summary points in this regard to the “Discussion and Conclusions” section.

To answer the question, slower-aging assumption for carbonaceous aerosols (BC and POM) is the main feature in MAM7 affecting BC distributions, as we demonstrated in the paper. However, there are other differences from MAM3, as we briefly summarized in the “Model Description” section (more details in Liu et al., 2012). MAM7 has more and separate size modes for sea salt and dust, and it has an explicit treatment of ammonia and ammonium.

- Section 3.5: On page 342 lines 14-15, it is said that “Note that the change does not directly impact the model’s convective cloud parameterizations involving heat, moisture and momentum”. However the indirect effects, which are already included in the simulation, appear to have substantial impact. Is there a way the Authors can try to distinguish between the direct and indirect processes?

AR: The change to the convective cloud processing of aerosols (i.e., wet removal and vertical transport) directly modifies aerosol distributions, but does not directly modify temperature or moisture. Changes to aerosol distributions lead to changes in aerosol direct radiative forcing, and changes to stratiform clouds (indirect effects) and thus temperature, moisture, and stratiform cloud radiative forcing. These changes to the atmospheric state can then produce changes in convective clouds in the model. Note that these changes to convective clouds are not the same as what would normally be considered aerosol indirect effects in convective clouds.

As shown in Fig. 9 and discussed in the text, the model change improves aerosol distribution but doesn’t degrade the model cloud and climate. It turns out that the change also improves various aspects of model climate. One of them is the reduction of low bias in LWP. We did attempt to gain more insight into how exactly the change affects LWP (see section 3.5), and found out that 75% of the increase is in stratiform clouds (primarily through aerosol indirect effects) with the rest of 25% increase from

convective clouds. Deeper process-level insight would require more carefully design of new simulations, which is not the scope of this study. We meant to stay focused on improving global aerosol distribution.

- Section 4: Finally, there are still some differences between the observed and simulated aerosols in the Arctic at the surface. Could the Authors try to elaborate a bit on why this might be the case?

AR: As we discussed already in Section 4, in addition to possible measurement uncertainties, uncertainties in emissions (a factor of 2-4 for BC; Bond et al., 2004) might explain the most part of the discrepancy, although there is likely more room for improvement in transport related processes (including wet removal) in the model. Further improvement to model processes would require more wet-removal-process-oriented measurements and understanding, which is currently lacking. Our sensitivity tests (in the Supplementary Material) show that not only global emission rates but also the regional distribution of emissions can strongly impact the Arctic surface aerosol concentrations. Some previous studies (e.g., Wang et al., 2011) have shown that emissions from particular regions would have to be doubled for models to match the observed surface concentrations in the Arctic. We have ongoing source-attribution studies to quantify importance of regional/sector emissions affecting the Arctic BC. We have also explored the dependence of aerosol transport on model resolution and meteorology/circulation, and this was found to be of limited importance climatologically, although quite important in specific cases (Ma et al., 2013a, b). We prefer not to speculate beyond what we have learned from the present study and the others mentioned above.

Technical/minor comments:

-Figure 6, legend: Please indicate "Observations (dotted black lines. . ..). A minor comment also in terms of how the model was sampled: does that make sense to sample the model along flight tracks using a 10-year January mean? Would that not be more

representative to use a model mean over the latitude-longitude range of each panel?

AR:

We have added "Observations" to the figure legend in Figures 6, 7 and 8.

The 10-year January mean really represents the present-day (year 2000) January mean because we are using the year 2000 emission inventory and year 2000 sea surface temperature. Regarding the sampling strategy, the way we did closely follows the treatment of observations and the original model-observation comparison by Schwarz et al. (2010). We believe it's better than the average in the entire latitude-longitude range because there is substantial spatial variability near emission source regions. Also, we actually tried calculating the model profiles both ways, and they differences were generally rather small.

-Figure S1: It is not easy to see the difference in emissions with the log scale.

AR: We have replaced the figure with a new one in linear scale for emissions.

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