We are grateful to the evaluation of the reviewers, which have allowed us to improve and clarify the manuscript. Below we address the review comments, with reviewer comments in italics and our response is in bold.

Two changes were made in the revised manuscript that were not in response to reviewer comments:

1. We replaced the model results at 4x5 horizontal resolution in the original manuscript with 34-month model results at 1.9x2.5 horizontal resolution. The change in the resolution makes the current manuscript serve as a good reference for future studies that are and will also be based on the 1.9x2.5 horizontal resolution (e.g., Wang et al., 2011). There are also a couple of other changes in the new simulation. First, the new simulation used the same oxidant concentrations and aerosol emissions as in the standard CAM5 release. In the original manuscript simulation, we used slightly different aerosol emissions and oxidants from a developmental version of CAM5. The main difference in the oxidants was higher NO_3 in the earlier version which led to a shorter DMS lifetime (0.56 days). Second, dust and sea salt emissions are tuned in the new simulation so that dust and sea salt burdens match those in the standard CAM5. The additional changes eliminate differences in anthropogenic emissions and minimize differences in dust and sea salt aerosols between the MMF and CAM5, which will facilitate the comparison of the two models in future studies. The simulation changes (resolution, duration, oxidants, and emissions) do not significantly alter the main message and conclusions of the manuscript.

2. Figure 17 is added to compare modeled BC vertical profiles with observations from the HIPPO campaign (Schwarz et al., 2010).

Anonymous Referee #1

Received and published: 2 December 2010

The paper presents a very interesting model tools that include small scale processes, e.g. convective clouds within a coarse model framework, which is certainly relevant for addressing scientific questions within the scope of EGU. Although the model tools are not new the combination represent a substantial advance in modelling science. The presentation is well structured, but with some deficiencies in the description of the methods and interpretations of results.

The title and abstract reflects the content of the paper although I would prefer to replace the wording "reasonable well" with a quantity at least once, e.g. within a factor of two.

We followed the reviewer's suggestion and now present the evaluation results

in a more quantitative manner in both the abstract and main text.

The methods are generally well outlined, but the distinction between convective clouds and "stratiform" clouds are somewhat unclear. If the methods are the same, how do you differentiate? The relative importance of convective scavenging is lower in this paper compared to earlier paper by some of the co-authors. Is this due to the speciation of convective or stratiform clouds?

In the ECPP approach (sect. 2.3), convective clouds are clouds in updraft and downdraft classes, and stratiform clouds are clouds in quiescent classes. We clarified this in the 1st paragraph in Sect. 2.3. The text now reads: "The ECPP approach uses statistics of cloud properties resolved by the CRM (Explicit-Cloud) to drive aerosol and chemical processing by clouds on the GCM grids (Parameterized-Pollutant), which allows us to explicitly account for the effects of both stratiform clouds (i.e., clouds in quiescent classes as defined in Sect. 2.3.1) and convective clouds (i.e., clouds in updraft and downdraft classes as defined in Sect. 2.3.1) on aerosols while being computationally feasible.". **Updraft, downdraft and quiescent classes are defined according to vertical velocities, hydrometeor mixing ratios, and precipitation rates, as described in details in Sect. 2.3.**

In the revised manuscript, we also compare the contribution of convective clouds to the total wet scavenging in the MMF model to those in CAM5 in Sect. 5. The text in Sect. 5 now reads(page 29, paragraph 3): "The smaller mass fraction located above 5 km in CAM5 is consistent with its smaller contributions of convective clouds to the total wet scavenging, which range 35-42%, though it is noted that the convective clouds is diagnosed in the MMF model by using CRM cloud statistics, while they are from shallow and deep convective clouds parameterizations in CAM5.".

Also I think the authors may be more precise in their description of the wet-scavenging process, in particular since it is emphasized a lot in model-model and model-measurement chapter. I assume that the in-cloud scavenging is modelled explicitly by the activated aerosol fraction?

Yes, the in-cloud scavenging is modeled explicitly by the activated aerosol fraction. This is described in detail in section 2.3.2 (See P. 13, paragraph 1). $f_{act\text{-}vert}$, and $f_{act\text{-}ent}$ are the activation fraction associated with vertical transport and entrainment, respectively.

There are also two other assumptions I would like the authors to comment on. Is the results sensitive to the assumed minimum grid vertical velocity of 0.1 m s-1 and the sub-grid vertical velocity minimum of 0.2 m s-1.

We chose these two minimum vertical velocities based on other studies, and we now comment on these two assumptions in the text. One comment was added in Section 2.2 about the minimum vertical velocity of 0.1 m s^{-1} used for calculating droplet activation in the CRM component, and the text now reads (P. 8, paragraph 1): "A minimum vertical velocity of 0.1 m s^{-1} is set for calculating aerosol activation, following Ghan et al. (1997) and Morrison et al. (2005). Sensitivity tests at a coarse GCM horizontal resolution (4°x5°) show that using a

minimum vertical velocity of 0.01 m s^{-1} instead of 0.1 m s^{-1} has little effect on simulated aerosol concentrations, since the aerosol activation in the CRM is only used for droplet formation and is not directly linked with the aerosol wet scavenging (which is treated in the ECPP, see details in Sect. 2.3.3)". **The other was added in Section 2.3.3 about the minimum vertical velocity of 0.2 m s^{-1} used for calculating droplet activation in the ECPP, and the text now reads (P. 13, the last paragraph): "**A lower bound of 0.20 m s^{-1} is used for the subgrid vertical velocity, the same as that used in the standard CAM5. Since most aerosol mass is activated for a vertical velocity larger than 0.1 m s^{-1} (except where accumulation-mode number concentrations are quite high), decreasing the minimum vertical velocity from 0.2 m s^{-1} to 0.1 m s^{-1} would have limited effect on simulated aerosol mass, though its effect on aerosol number concentrations would be larger (Jensen and Charlson, 1984; Abdul-Razzak and Ghan, 2000). We note that the minimum vertical velocity used for droplet activation in GCMs is not well constrained yet.".

The set-up of the experiments are concisely described, although personally I think 2 months of spin-up is short wrt polar regions, in particular over the Antarctic regions. I do not think it will change any of the conclusions though.

In the revised manuscript, we now use 34 months of model results at 1.9x2.5 degrees horizontal resolution.

As mentioned above I think the description of scavenging is a weak point. In particular this is the case when it comes to interpreting the budgets, and to some minor extent comparison with measurement. The authors define a wet removal rate coefficient by the inverse of the residence time. This bulk parameter combine a lot of the physical properties in the model, including updraft velocity, activation rates, cloud volume, moisture convergence precipitation frequency ..... I assume and understand that the authors would like to include the activation budget together with cdnc calculation and in-direct effect, but I nevertheless ask the authors to consider including numbers for scavenging coefficients as calculated by the activation. This may also provide information on whether the relatively low scavenging is due to the activation or the precipitation distribution.

We agree with the reviewer's comments that the wet removal rate coefficient is a bulk parameter, and it would be desirable to include scavenging coefficients as calculated by the activation. However, we focus on the comparison with the AeroCom models in this manuscript. Since the model results from the AeroCom models did not include the activation fraction, adding the activation fraction from the MMF would not strengthen this comparison. What is more, the MMF model did not include this information in its output. Given the expensive nature of the model, it is not possible for us to rerun the simulations to get this information. In the future, we plan to examine differences in simulated aerosol fields in both the MMF and CAM5. Wet scavenging will be the focus, and the scavenging coefficient parameter will be examined. We will run the models nudged with reanalysis winds so that the effects of large-scale transport will be separated from the effects of wet scavenging and convective transport.

On the other hand I am wondering whether the ccn part of the paper should be included in-direct effect paper, since I find the sometimes large differences between ccn and cdnc interesting in the context of activation more than wrt to theoretical measurements.

We retained the CCN results, since CCN (at appropriate supersaturations) is the measure of aerosol number that is the most relevant cloud droplet number concentrations. We included a comparison of the CCN concentrations between the MMF model and CAM5 in a separate manuscript that examines aerosol indirect effects (Wang et al., 2011).

Some details.
I assume "hydrometers" should be "hydrometeors" (found several places throughout the paper)
Corrected.

1629, line 22 "addressed" instead of "address"?
Corrected.

1630 last line "those" –> "these" ?
Corrected.

1634 cloud-borne vs interstital. cloud-borne in this connection is inside cloud droplets, not all particle within the cloud volume?
Yes. We clarify this by replacing 'cloud borne (in-cloud)' with 'cloud borne (inside cloud droplets)'.

1643 Martensson should be Mårtensson
Corrected.

1646 Long lifetime of sea-salt. Is the gravitational settling 3 dimensional and take into account hygroscopic growth?
Yes, the gravitational settling is 3-dimensional and takes account of hygroscopic growth. This is described in Liu et al. (2011, in preparation). The sea-salt lifetime is also rather long in the standard CAM5 (0.90 days). In the modal approach, the settling and dry deposition velocities are quite sensitive to the mode standard deviation , and using a single coarse mode with standard deviation = 1.8 contributes to the longer lifetime.

1650 Elevated so2 layers over the Pacific. The emission data-set includes a number of volcanoes. The location of active volcanoes will of course vary from year to year so the difference in the position of elevated so2 layers may be caused by variation in volcanic activity.
We agree. The volcanic SO2 emissions from Dentener et al. (2006) included both continuously degassing and explosive volcanoes. However, we used only the continuously degassing emissions, which are about six times greater

(global total) than the explosive emissions. We added one more comment about the elevated SO2 over Hawaiian is added in Section 4.1(P. 22, 1st paragraph): ".An elevated SO_2 layer is simulated in the lower troposphere (at about 3-4 km) near Hawaii, which comes from the SO_2 emissions from Hawaiian volcanoes.".

1651-14 sties –> sites
Corrected.

1653-10 middle –> mid
Corrected.

1654-22 Aikten –> Aitken
Corrected.

1656-22 strong —> high ?
Replaced 'strong' with 'high'.

1668 Table 1 lifetime range 0.2-2.6 should be 0.6-2.6 ? At least that is what is said in the text
That is correct, and fixed.

1695 figure 22: Close to impossible to distinguish between 0.4 and 0.6.
The colors were changed.

Anonymous Referee #2

Received and published: 30 December 2010

General Comments

This paper describes improvements made to the "Multi-scale Modelling Framework" (MMF) which essentially modifies a global climate model to have its cloud properties and processes to be as simulated by an embedded cloud-resolving model rather than by conventional parameterizations.

The paper describes three main improvements on the "original MMF framework" (as in Khairoutdinov et al, 2008) in CAM v3.5: i) the use of a two-moment modal aerosol scheme in CAM, ii) the use of the explicit-cloud parameterized-pollutant (ECPP) approach to use the CRM-scale cloud information to affect the gas/aerosol simulated on the climate model scale, iii) the use of a two-moment cloud microphysics scheme in the CRM rather than the original single-moment scheme.

This improved version (referred to as the PNNL-MMF) represents a novel and promising way to use the MMF "super-parameterization" approach to improve the representation of aerosol-cloud interactions in global climate models.

As well as clearly describing the developments to the CAM model, the paper provides a

comprehensive documentation of how the global aerosol simulated by the improved model compares against other global models and evaluates a number of key quantities against an impressive collection of key observational datasets suitable for assessing global aerosol models. The description of the improved MMF implementation within CAM is interesting and the evaluation of the improved CAM is comprehensive and, as such, the paper is certainly suitable for publication in Geoscientific Model Development. The paper is in good shape and reads well throughout. The introduction is appropriate and the model description is comprehensive and clearly explained.

My main criticism however, is that, although the paper presents evaluation of the improved model against an impressive number of observational datasets, there is no explanation of how much better the improved CAM is compared to the current standard CAM5 version without the MMF approach. The final sentence of the abstract states that "the MMF version of CAM5 simulates aerosol fields as well as conventional aerosol models". I guess one would hope that it might simulate the aerosol better or more realistically. Here, and elsewhere in the paper, the benefits of the multi-scale approach needs to be stated more clearly.

For instance, are the PNNL-MMF-CAM5 simulated size distributions and size-resolved number concentrations in Figures 17-20 better or worse than those with the standard CAM5?

Without this information, the reader can only see this new framework as a whole without being able to assess whether the use of the CRM information improves the model or not. Or which aspects of the model are improved or made more realistic by this new approach.

From the description in the paper, the Liu et al (in prep, 2010) paper will describe runs of the standard CAM5 model that could be included in this analysis and used to specifically examine where the use of cloud-resolving scale statistics changes the model predictions. If it is straightforward to do, I would ask the authors to consider adding this standard CAM5 simulation as a reference model line in as many of the figures as possible — this ought to be possible for many of the Figures and would greatly improve the paper, and help to understand the impact of the new approach on the simulated aerosol properties.

We appreciate the reviewer's suggestion about adding CAM5 results to the current manuscript. We agree with the reviewer that it is an important scientific task to examine differences in simulated aerosols fields in the PNNL-MMF and CAM5. Efforts on this task may lead to new insights about the aerosol treatment in conventional aerosol-climate models, which may eventually help to improve the treatment in conventional models. However, we feel that this task is out of the scope of current manuscript, and will pursue it in future studies.

The objective of the current manuscript is to document the new model and to evaluate its performance in simulating aerosols and sulfur gases. We compared simulated aerosol budgets with the AeroCom models and simulated aerosol parameters with observations, which led us to conclude that "the MMF

version of the CAM5 simulates aerosol fields as well as conventional aerosol models**". It is not an objective of the current manuscript to show how the PNNL-MMF may improve aerosol simulations compared with CAM5. Though the PNNL-MMF model is based on CAM5, all turbulence and moisture parameterizations are replaced by the embedded CRM. Given the differences in simulated clouds and precipitation, comparing simulated aerosols and understanding the differences between the two models is a substantial effort that is beyond the scope of the current manuscript. Nonetheless, we have added some discussion of differences between the MMF and CAM5 results in Section 5.**

In a separate aerosol indirect effect manuscript (Wang et al., 2011), we do show some comparisons between CAM5 and MMF (see Figs. 7-10 in Wang et al., 2011). Fig. 7 in Wang et al. (2011), which reproduces Fig. 18 of this manuscript, shows that the PNNL-MMF and CAM5 produce similar aerosol size distributions, and the PNNL-MMF agrees slightly better with observations. Simulated BC seasonal cycles in the MMF and CAM5 in the polar regions are compared with observations in Fig. 8 in Wang et al. (2011), and it shows that the MMF agrees better with observations in terms of both magnitude and seasonal cycles. In the future, we plan to further explore why the MMF produces better aerosol fields in the Arctic regions, focusing on the treatment of wet scavenging. We plan to run both the MMF and CAM5 nudged with reanalysis wind fields, which will help us to isolate the effects of large-scale transport from the effects of wet scavenging and convective transport.

Another important concern for us is that the manuscript describing the standard CAM5 aerosol treatment (Liu et al., 2011, in preparation) is still in preparation, so the current manuscript will most likely be published first. As many observational datasets used in the current manuscript are also used to evaluate the standard CAM5, including CAM5 results on many of our figures would preempt the CAM5 paper. As the CAM5 aerosol treatment involves many people and groups, we do not want to see this happen.

Given these concerns, we have included only limited CAM5 results in the current manuscript, and we will further explore differences in simulated aerosol fields between CAM5 and the MMF model in the future.

Another aspect of the paper that needs improving is that, although in some of the comparisons to observations (Figures 7-14), there are values for the correlation coefficient R given, in many of the Figures there are no statistical measures of the model comparison to the observations at all (e.g. Figure 21).

Also, even in the Figures which do have R values, I recommend that there should also be added a measure of the model normalised-mean-bias or standard-error since just the R value does not constrain the skill of the model very well.

I recommend that an extra Table (or perhaps 2) be added to the paper which give the R and normalised-mean-bias/error values for each of the observational datasets used to assess the skill of the model.

Done. We have added correlation coefficients and normalized-mean-biases to the model-observation comparison figures (Fig. 7-21).

Related to this, I also recommend that, if possible, R and bias/error values are given for the standard CAM5 simulation from Liu et al (in prep, 2010) and then the reader can see exactly how the model has improved or otherwise with the incorporation of the new MMF development.

See our reply above. The comparison between the MMF and CAM5 will be the topic of future studies.

However, overall this is a nice paper and I recommend it is published once the issues I have raised have been sufficient addressed.

Specific Comments

1) Abstract – the 2nd part of the abstract which summarizes the skill of the model uses several times the phrase "are in reasonable agreement" – the authors should give some kind of quantitative measure here in each case in terms of correlation and/or mean bias values.

We now present the evaluation results in a more quantitative manner in both the abstract and main text.

2) Introduction – pg 1629, line 9 – the sentence "The MMF models have been shown to improve climate simulations in several important ways (...several refs...) " should be rewritten. State very briefly exactly they key ways that the climate models are improved by using the approach.

Additional information was added, and the text now it reads(P. 4, Paragraph 2): "The MMF models have been shown to improve climate simulations in several important ways, including representation of convective clouds, the diurnal cycle of precipitation, and the subseasonal variability of tropical climate associated with the Madden–Julian oscillation (MJO) and equatorially trapped waves (Ovtchinnikov et al., 2006; Khairoutdinov et al., 2008; Pritchard and Somerville, 2009a, b; Tao et al., 2009)"

3) Section 2.1 – pg 1631, lines 23-24 – please explain what mechanism(s) are used to represent Aerosol nucleation in the model. Since the paper includes several comparisons to observations of the size distribution in section 4.2 this should be explained here. On page 1653 there is reference to a boundary layer nucleation mechanism being included in the model – but this is not described anywhere in the paper – please include a sentence in section 2.1 on which binary nucleation mechanism is used and the approach (and coefficients) used for boundary layer nucleation.

Done. The following sentence was added (P. 6, the last paragraph): "Aerosol nucleation from H_2SO_4 is treated and includes binary H_2SO_4-H_2O homogeneous nucleation, based on the parameterization of Vehkamaki et al. (2002), and empirical boundary layer nucleation, based on the first order nucleation rate in H_2SO_4 from Sihto et al. (2006) with a first order rate coefficient of 1.0×10^{-6} s^{-1} as in Wang et al. (2009). The new particles are added to the Aitken mode, and the parameterization

of Kerminen and Kulmala (2002) is used to account for the loss of the new particles by coagulation as they grow from critical cluster size to Aitken mode".

4) Section 2.4, pg 1643, lines 3-9: Here, and at several other points in the paper, the phrase ""Gas phase SOA" is used—and indeed an acronym SOAG is used to describe this. I would suggest the authors not use the phrase "Gas phase SOA". Although one could argue that technically, since the term "aerosol" represents both the particle and gas-phase, the phrase "gas-phase SOA" does make sense, I feel this terminology is confusing for the reader because it suggests that SOAG is the gaseous part of a semivolatile aerosol, whereas in fact all the "SOAG" is condensing into the particle phase. I would suggest that the authors remove reference to "SOAG" and instead either refer to the gas with a different name reflecting the fact that it is an condensing gas phase organic species (e.g. CONDORG) or else re-phrase so that there is no reference to the SOA in the gas phase (and remove the top-part of Table 4 which refers to SOAG) except in section 2.4 where it should be explained that this is the technique for producing SOA in the model.

The terms "SOAG" and "gas-phase SOA" have been replaced by "condensing gas-phase organic species". Results for and discussion of the SOAG burden and lifetime were removed from Table 4 and Section 3.1. Additional information was added in section 2.4 about the SOA treatment in the CAM5, and the text now reads (P. 15, Paragraph 2): "In the CAM5 simplified SOA mechanism (Liu et al., 2011, in preparation), condensing gas-phase organic species, which condense (reversibly) to give SOA, are emitted directly in the model using prescribed yields for several primary VOC classes, rather than being formed by atmospheric.".

5) Section 3.1 – pg 1644, lines 24-25: the text refers to the larger fraction of the sulphate burden above 5km than other AEROCOM models. To what extent is this a product of the MMF approach for the scavenging and to what extent is it a general feature of the CAM model (i.e. without the MMF approach)? There should be some reference here to the values in the standard CAM model – again the reader needs to have a better handle as to how the MMF-CAM framework compares with standard CAM – can values from Liu et al (in prep., 2010) be added to Tables 1-6? Also this is referred to in the Summary (page 1658, lines 24-26) and it needs to be much clearer in the paper whether this is a problem with the way the MMF approach has been implemented or a general problem with CAM5.

As we mentioned above, though the PNNL-MMF model is based on CAM5, all turbulence and moisture parameterizations are replaced by the embedded CRM, which leads to different cloud and precipitation fields and different cloud processing of aerosols. We feel that it is difficult to use the term 'general problem' for some aspect of the simulated aerosol fields in the MMF and CAM5 unless that aspect can be attributed to common features of the two models (e.g., emissions, modal representation, dynamical core, surface models). The high sulfate burden above 5 km is more likely due to model features that differ (clouds and precipitation). Also, as discussed above, we have several reasons for not including MMF-CAM5 comparisons in this manuscript. Nevertheless, we added a paragraph in Section 5 that discusses differences in

above 5 km burdens between the MMF and CAM5. That paragraph reads (P. 27, paragraph 3): "We note that the mass fractions of aerosols located above 5 km in the standard CAM5 are smaller than those in the MMF model. The mass fractions of sulfate, BC, POM located above 5 km in the standard CAM5 are 30%, 20%, 20%, respectively, and are close to those in the AeroCom models. The smaller mass fraction located above 5 km in CAM5 is consistent with its smaller contributions of convective clouds to the total wet scavenging, which range 35-42%, though it is noted that the convective clouds is diagnosed in the MMF model by using CRM cloud statistics, while they are from shallow and deep convective clouds parameterizations in CAM5. Differences in convective transport, wet scavenging in stratiform and convective clouds, and long range transport between the MMF model and CAM5 may lead to these differences. Further studies are needed to identify the causes for the differences between the CAM5 and MMF**." We also added a note in the 3rd paragraph of sect. 3.1:** "(see discussions in Sect. 5 about the differences between the MMF and CAM5)**".**

6) Section 4.2 – pg 1654 lines 3-5 – the authors attribute the "difficulties in simulating the monomodal size distributions in the free troposphere" to "the modal representation of the aerosol size distribution in the MMF model". The authors should clarify what is meant here. Firstly I presume they are referring to the treatment of the aerosol in CAM5. If so then they should state that it is in that model rather than "the MMF model" as it is what is used in CAM5. Secondly, there are different implementations of a "modal" aerosol in different models – for instance Stier et al (2005) and Mann et al (2010) use 7 modes which include a separate nucleation mode (representing particles smaller than 10nm diameter) in addition to an "Aitken mode" to represent particles in the 10nm-100nm size range. In this study, only 3 modes are used with only 1 mode representing particles across both these size ranges. Indeed the authors explain in section 2.1 page 1631 lines 13-21 how they use the 3-mode approach rather than a 7-mode approach for computational efficiency reasons. To what extent are the deficiencies here a product of the use of the simplified (3-mode) modal approach rather than the detailed (7-mode) approach and to what extent is it a problem with all modal approaches using constant mode-widths? I realize the authors may not be able to answer that question in the revised paper but there should be reference to the different possible cause here and that it may not be a general problem with the modal approach, which may be inferred by the reader here. Again, reference to how well the standard CAM5 simulation performs against these observations would help here.

We mean modal approach in general here. CAM5 and other models with modal approaches perform similarly in simulating the aerosol size distributions in the lower and middle free troposphere. We have clarified our argument in the text. The fixed standard deviations and the fixed boundary between the Aitken and accumulation modes likely contribute to the difficulty in simulating a monomodal size distribution in the lower and middle troposphere. An additional nucleation mode may have little impact on this, since the same problem is seen in Stier et al. (2005). The revised discussion was moved to section 5, and the text now reads (P. 30, paragraph 2): "The difficulties in

simulating the monomodal size distributions in the lower and middle free troposphere are also true for the standard CAM5 (not shown), and the CAM-IMPACT model (Wang et al., 2009). The MMF and CAM5 also tend to simulate too much bimodality in the boundary layer over some continental regions (not shown), as do CAM-IMPACT and the ECHAM5-HAM model (Stier et al., 2005). Most modal aerosol approaches represent aerosol size distributions by using several log-normal modes with fixed standard deviations and fixed modal boundaries. The fixed modal boundaries limit their capability to simulate monomodal (or near-monomodal) aerosol size distributions when the single mode wants to be centered near the boundary of the Aitken and accumulation modes. As condensational growth causes the Aitken mode to grow closer to this boundary, the mode merging (or remapping) algorithm transfers part of the Aitken mode into the accumulation mode, maintaining the bimodal distribution"**.**

7) Section 5 – pg 1658 lines 15-22 – the authors refer to the accumulation mode being underestimated and the Aitken mode being overestimated in the free troposphere. The only possible cause given in this section is that the SO2 is over-estimated. Isn't is also possible that the scavenging approach being implemented here may be the cause? Or couldn't it also be possible that the simplified 3-mode treatment of the aerosol is causing problems representing nucleation with only 1 mode covering sub-100nm particles? Also, as referred to in the last part of the section 5 on pg 1659 (lines 4-10), could the fact that the low cloud are biased be affecting the processing of Aitken mode particles into the accumulation mode? These (and any other) possible causes for this bias should be mentioned in this section of the conclusions rather than suggesting it is likely only a problem with the SO2.

We revised this part of Section 5, adding comparison with CAM5 results, and listing several possible reasons for the overestimation of both Aitken and accumulation mode particles in the upper troposphere over the mid-latitudes. The text now reads (P. 30, the last paragraph): "Simulated accumulation mode aerosol number concentrations are overestimated in the middle and upper troposphere over the mid-latitudes, while Aitken mode aerosol number concentrations are overestimated in the upper free troposphere, in comparison to the INCA campaign measurements (Fig. 20). The overestimation of accumulation mode number concentrations in the middle and upper troposphere is consistent with the large aerosol mass fraction above 5 km in the MMF model compared with the AeroCom models, and the overestimation of Aitken mode number concentrations is consistent with the excessive SO_2 concentrations in the upper troposphere. We note that the standard CAM5 simulates less Aitken and accumulation mode aerosol number concentrations in the middle and upper free troposphere over the same locations and agrees better with observations (not shown). This can be caused by differences in simulated convective transport, wet scavenging, and cloud fields in both the MMF and CAM5. **".**

Minor Comments & Typos

1) Abstract – pg 1626 lines 5,6 and afterwards: "Global Climate Models (GCMs)" – the acronym GCM is generally accepted to refer to "General Circulation Model" rather than "Global Climate Model". I suggest that, if the authors are specifically referring to the climate model, they avoid the GCM acronym as it can be confusing to the reader.

Thanks. We do mean " general circulation model" here, so we replaced all occurrences of "global climate model" with "general circulation model".

2) Section 3.1 – pg 1643, lines 23-24 – Sentence beginning "For gas species, a range of results from other models. . …" mentions Liu et al (2005) twice in succession – and so does the caption to Table 1 – suggest just to state "values listed in Liu et al (2005)" – for a while I thought it must be a typo but I now see what you mean here – but suggest to reword this.

Corrected.

3) Section 4.2 – pg 1652, lines 25-28 – The text explains that the model data is sampled over the same regions as the 15x15 degree gridded observational data – but there is no explanation of the temporal sampling here – is this an annual mean

We clarified this in the caption of Figure 17, which now reads: "Annual-mean model data is sampled over the low latitudes (30°S-30°N). Over the mid- and high latitudes, the model data are sampled in summer (December to February for the latitude bands of 75°S-45°S, November to March for the latitude bands of 45°S-30°S, May to September for the latitude band of 30°N-45°N, June to August for the latitude bands of 45°N-90°N).".

4) Section 4.2 – pg 1653, line 3 and pg 1654 line 22 and pg 1658 line 19– Aikten à Aitken.

Corrected.

5) Section 4.2 – pg 1653, lines 22-23 – the authors refer to the "depletion of accumulation mode particles in the boundary layer" – I presume the authors are referring here to scavenging by wet removal – please clarify what is meant by "depletion".

Yes, the depletion is by wet removal. We clarified this in the text, which now reads (P. 26, paragraph 1): "The observed monomodal size distribution in the free troposphere is believed to result from the wet removal of accumulation mode particles in the boundary layer, and the generation of Aitken particles from nucleation in the free troposphere (Raes et al., 2000)**.".**

Reference:

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