The authors thank the referee for the constructive comments which help to improve the manuscript. Our point-to-point replies to the comments (in blue) are given below (the original comments are copied here in *Italic*). The manuscript has been revised accordingly.

Anonymous Referee #2

Summary

The authors report on a study conducted with a global climate model to which a new representation of aerosol nucleation from the gas phase was added. Effects of aerosol nucleation involving neutral and charged sulfuric acid molecules on aerosol concentrations, cloud properties, radiation, and precipitation are investigated. The manuscript is well written and the results are interesting, relevant, and worth publishing. On the negative side, I see some fundamental issues regarding the identification of aerosol-cloud effects in climate models, which I explain below, along with some other points. I would like to ask the authors to rebut my arguments or to resolve the issues in the manuscript prior to publication. In particular I would strongly encourage a discussion of the limitations in identifying aerosol-cloud effects in climate models in the manuscript, and some comparison of aerosol properties (size distributions, vertical profiles of aerosol concentration) with observations prior to publication.

General comments

On the issue of identifying aerosol-cloud effects in climate models

To my knowledge, there is no dependable prognostic cloud fraction scheme that includes a dependence on cloud drop or aerosol number and cloud liquid water content for global/climate models. Such a scheme would be the straightforward representation of the cloud-lifetime or 2nd indirect aerosol effect (Albrecht, 1989). Schemes of this kind exist, but suffer from various problems and are currently not in regular use. Instead, if I read the NCAR technical note NCAR/TN-486+STR correctly, the stratus cloud fraction in CAM 5 is parameterized as a function of relative humidity only, while for other cloud types, the cloud fraction is parameterized using dynamics variables. These are established approaches in climate modeling. However, relative humidity depends, among others, on dynamics. The question then arises whether the change of cloud fraction at different aerosol nucleation levels reported in this work is only partially the manifestation of the cloud-lifetime or 2nd indirect aerosol effect (Albrecht, 1989), and to some extent the side effect of dynamics responding the the concurrent change in cloud albedo via the 1st indirect aerosol effect (Twomey, 1991). The Twomey (1991) and Albrecht (1989) effects can be better identified with models that can impose the same large scale dynamics on two different simulations, e.g. by a data assimilation procedure such as nudging, although this does not entirely eliminate the problem, because it could remain on scales that are not affected by the nudging procedure. This highlights the issue in identifying aerosol-cloud effects in climate models. Specifically with the present work in mind, because the dynamics in runs with different aerosol nucleation levels can be different, it seems that the response of cloud cover to different aerosol nucleation levels cannot be unambiguously identified as the cloud lifetime or 2nd indirect effect Albrecht (1989). I would like to ask the authors to discuss this in the manuscript.

In the model simulations reported here we are using the prescribed sea surface temperatures in different simulations. This eliminates the “slow” responses of the climate system through air-sea

interactions due to the aerosol perturbations. We are aware that this does not remove all dynamics response. The shortwave and longwave cloud forcing changes due to different aerosol nucleation levels reported in this paper include aerosol 1st and 2nd indirect effects and semi-direct effect of absorbing aerosols. Although it is difficult to separate dynamical effects from microphysical effects on cloud cover, Ghan et al. (2012) separate aerosol semi-direct effects from microphysical effects on shortwave cloud cover (which depends on liquid water path as well as cloud fraction), and find that semi-direct effects are small. We have added this discussion to the manuscript.

Internal variability

Without a data assimilation procedure such as nudging, different dynamics between two simulations can arise because of internal variability of the model. The different dynamics needs not to be caused causally by a difference in a specific process, and the differences average out over sufficiently long simulations. In simulations that are not sufficiently long, however, differences from internal variability may not average out. Does the averaging over 5 years of simulation sufficiently reduce the effect of internal variability and isolate the signal that is caused by the different aerosol nucleation levels?

From our previous experiences with CAM5 with prescribed sea surface temperatures, the indirect effect from the 5 years of simulations are very similar to that from the 10 years of simulations (the global mean difference is less than 0.1 W m^{-2}). 5 years of simulations are sufficient to smooth out the internal variability (Meskhidze et al., 2011; Gantt et al., 2012; Gettelman et al., in press, 2012). Ghan et al. (2012) find the standard error based on 5-years of simulation in the global mean aerosol indirect effect to be about 0.1 W m^{-2}, which is far smaller than the shortwave cloud forcing signal estimated in this study. We have used the same method to estimate the uncertainty, and have included in Table 1.

Comparison with observations

The authors have conducted a comparison/evaluation of their results with selected observations: Among the observations used are globally averaged liquid water path, cloud forcing, and cloud cover, and the spatial distribution of cloud cover and precipitation. With the exception of precipitation, the comparison of the results with the observations would, however, appear to include circular reasoning to some extent, because global/climate models are usually tuned to reproduce some or all of the observed values. Hence under closer inspection, the comparison with observations would seem to provide less insight into the capabilities of the model than seen from afar. Since the present study addresses aerosol formation, I would like to very strongly encourage that a comparison with quantities related to aerosol, such as aerosol size distributions and vertical profiles (see, e.g., Zhang et al., 2010; Kazil et al., 2010) is done by the authors before the manuscript is published. The comparison/evaluation of model results with observations can provide valuable hints to future model development and improvement. The comparison would therefore be very beneficial for this work and for future work with CAM. However, I do not think that to publish a model paper, it is required that the results agree with the observations perfectly, or even imperfectly.

We agree with the referee that model evaluations with observations are important. The measurements of aerosol size distributions and vertical profiles are quite limited. Instead, we have

added evaluation of CCN concentrations which are directly relevant to the aerosol indirect radiative forcing. Also see our reply to a similar comment by Referee #1. We would like to point out that Liu et al. (2012) have done a lot of aerosol evaluation. CAM5-MAM is also part of AeroCom intercomparison of aerosol microphysics.

Detailed comments

Reviewer comment: *Please write in the abstract and in the introduction which molecules take part in ion-mediated nucleation (IMN) and in binary homogeneous nucleation (BHN).*

Done.

P.2/L.5: *Here we implement for the first time a physically-based treatment of IMN into the Community Atmosphere Model version 5.*
P.5/L.15: *In this study, we implement for the first time the IMN mechanism (Yu, 2010a) in CAM5.1.*
Reviewer comment: *Is it justified to write "for the first time"? Other groups have implemented aerosol nucleation from ions in climate models, using other schemes to describe the process. I do therefore not see that the present research is fundamentally different compared to other studies. The ion-mediated nucleation (IMN) scheme used in this work is one description of aerosol nucleation from ions among others, and CAM 5 is one climate model among others.*

To address the referee's concern, we have delete "for the first time" in the revised manuscript.

P.2/L.6: *... physically-based treatment of IMN ...*
P.5/L.16: *As mentioned earlier, the physically-based IMN ...*
Reviewer comment: *Please explain "physically-based" in the text. Since "physically-based" applies to the chosen method to describe aerosol nucleation from ions, it should not be left uncommented, if only to highlight the contrast to other methods of describing aerosol nucleation from ions.*

When we say that IMN is "physically-based", we mean that IMN is based on a kinetic nucleation model that solves explicitly the interactions of clusters of various sizes (from single molecules and critical clusters) and different types (positively and negatively charged as well as neutral). Details of the IMN scheme are given in Yu (2006, 2010). To avoid the confusion, we have deleted "physically-based" from the two sentences.

5.3/L.3: *The nucleated particles are added to the Aitken mode, with coagulation loss as they grow from critical cluster size to Aitken mode size taken into account following the parameterization of Kerminen and Kulmala (2002).*
Reviewer comment: *Placing nucleating particles directly into the Aitken mode, which usually covers particles in the size range of _25-70nm is not ideal, because Aitken mode particles can be activated, while nucleation mode particles are usually too small to be activated. To avoid resulting errors, it would be best to add a dedicated nucleation mode to the model. It seems excessive to request this for this work, but the manuscript should mention that this is something that needs to be done in the future.*

We agree that not treating the nucleation mode explicitly leads to less accurate simulation of the aerosol distribution. However, the impact on CCN is much less than on total particle number. (See our response to comment by to Reviewer 1.) Also, new particles placed in the Aitken mode (at 12 nm diameter) will generally not act as CCN until they have grown to larger sizes (e.g., 50 nm). We are currently implementing a version of the CAM5 modal aerosol module that includes a nucleation mode. Discussion of these points has been added to the revised manuscript.

Reviewer comment: *The analytical formula of Kerminen and Kulmala (2002) only applies when self-coagulation of the nucleating particles is negligible. In conditions where the nucleation rate is large, e.g. at high concentrations of nucleating molecules, or at very low temperatures (upper troposphere, polar regions), neglecting self-coagulation will lead to errors in the number of particles from nucleation that are placed in the Aitken mode. These errors will propagate into the effect of nucleation on aerosol concentrations, cloud properties, and radiative forcing. This limitation of the present study should be mentioned.*
Reviewer comment: *Furthermore, to avoid all these problems, why did the authors not use the more recent parameterization of Anttila et al. (2010), which does take into account self-coagulation of the nucleating particles? This parameterization was developed specifically with large scale models in mind, and would seem most appropriate for a study such as the present one.*

The new parameterization of Anttila et al. (2010) is an improvement on Kerminen and Kulmala (2002), but has not yet been incorporated into CAM5. We note, however, that the new parameterization still tends to over-predict new particle formation in comparison to the aerosol dynamics model (see average relative bias and error statistics in their Table 4) by amounts similar to the 2002 parameterization.

Reviewer comment: *The more advanced parameterization of Anttila et al. (2010) appears to be able to bridge growth of nucleating particles up to particle diameters of 3-10 nm (Anttila et al., 2010). When bridging a larger diameter range, the performance of the parameterization increasingly worsens. What is the size range in the present work that is bridged with the formula of Kerminen and Kulmala (2002)? Please provide this information in the manuscript, together with the particle size range covered by the Aitken mode. What is your assessment of the bridged size range in your work in the light of the information provided by Anttila et al. on their more advanced paramenterization, whose accuracy deteriorates for diameter ranges in excess of about 10 nm?*

The upper diameter used with the Kerminen and Kulmala parameterization is 12 nm, which is not much larger than the 10 nm value above which the parameterization performance deteriorates. The model does not specify an explicit size range for the Aitken mode. As noted in Liu et al. (2012), the Aitken mode size range, based on 10^{th} and 90^{th} percentiles of the global annual average number distribution, is 15-53 nm. Discussion of these points has been added to the revised manuscript.

P.5/L.29: *... except that, in order to clearly assess the effect of nucleation, the fraction of anthropogenic sulfur emitted as primary sulfate (used to represent sub-grid nucleation process) has been set to zero. Many previous global aerosol modeling studies have assumed some fraction (0-5 %) of anthropogenic sulfur emitted directly as sulfate particles to account for the new particle formation in sub-grid SO2 plumes (Luo and Yu, 2011; and references therein). However,*

to assume a constant fraction of sulfur emitted directly as particles (with an assumed percentage partitioning into Aitken and accumulation modes) may lead to large uncertainty in the simulated spatiotemporal distribution of particle number concentrations, owning to the strong dependence of sub-grid nucleation on many environmental parameters ...
Reviewer comment: *This reasoning does make sense up to a point: SO2 emission plumes take up a very small volume compared to the grid volumes of climate models. Letting the all anthropogenic sulfur being emitted as SO2 and letting aerosol nucleation proceed in the resulting conditions means extending (and diluting) the emission plumes over an entire grid box. At the resulution of 1.9_×2.5_ used in this study, correct aerosol concentrations from nucleation in subgrid-scale SO2 emission plumes should not be expected. Therefore, I think the approach chosen here does not guarantee that uncertainty in the results is reduced compared to the original approach where a given fraction of anthropogenic sulfur emissions is apportioned to primary sulfate aerosol particles. Please discuss this point in the manuscript text.*

Yes, we added some further discussion on this. Also see our reply to a similar comment by referee # 1.

P.7/L.1: *H2SO4 vapor from both anthropogenic (fossil fuels, etc.) and natural (DMS, volcano, etc.) sources is known to play an important role in forming and growing new particles. H2SO4 column burdens are high in the source and associated outflow regions (Fig. 1a, b),*
Reviewer comment: *The reader is left alone in identifying what type of source and associated outflow regions contribute to H2SO4 column burdens. It would be helpful to add a few sentences that say, e.g., that the high H2SO4 burdens extending from northern Chile to northern Argentina and southern Brazil are due to volcanic emissions in the Andes.*

We have added the following sentences:
For example, H2SO4 column burdens over East Asia, Europe, and Northern America are associated with anthropogenic emissions. The high H2SO4 burdens extending from northern Chile to northern Argentina and southern Brazil are due to volcanic emissions in the Andes.

P.7/L.6: *"Because of ion-dipole interaction ..."*
Reviewer comment: *Some additional information would help the reader: "Because of the attractive interaction between the HSO−4 ion and the electric dipole of H2SO4, ..."*

We have modified the sentence as suggested.

P.9/L.5: *"... there exist substantial differences between the predicted and observed global mean LWP and precipitation ...*
P.9/L.15: *As mentioned earlier, the present CAM5 does not consider indirect effect of aerosols on convective clouds which dominate precipitations. Further research is needed to understand the interaction of aerosols with convective clouds and improve the representation of such interaction in CAM5."*
Reviewer comment: *Is it possible that some of the LWP and precipitation biases in the results could also arise from the specific tuning of subgrid-scale processes/quantities of the model, such as autoconversion, entrainement, and deep cumulus cloud fraction in CAM and not just from the fact that the model does not include aerosol effects on convective clouds?*

Yes, some of the LWP and precipitation biases can result from the representations of other physical processes such as cloud microphysics, macrophysics and convection, not just from the fact that the model does not include aerosol effects on convective clouds. We have made this clarification in the revision.

References

Anttila, T., V.-M. Kerminen, and K. E. J. Lehtinen: Parameterizing the formation rate of new particles: The effect of nuclei self-coagulation, J. Aerosol Sci., 41, 621-636, 2010.

Gantt, B., J. Xu, N. Meskhidze, Y. Zhang, A. Nenes, S. J. Ghan, X. Liu, R. C. Easter, and R. Zaveri: Global distribution and climate forcing of marine carbonaceous aerosol. 2. Effects on cloud properties and radiative forcing. Atmospheric Chemistry and Physics, 12, 6555–6563, doi: 10.5194/acp-12-6555-2012, 2012.

Gettelman A., X. Liu, D. Barahona, U. Lohmann, and C. Chen, Climate Impacts of Ice Nucleation, Journal of Geophysical Research, in press, 2012.

Ghan, S. J., X. Liu, R. C. Easter, R. Zaveri, P. J. Rasch, J.-H. Yoon, and. B. Eaton: Toward a minimal representation of aerosols in climate models: Comparative decomposition of aerosol direct, semi-direct and indirect radiative forcing. J. Climate, 25, 6461-6476, doi: 10.1175/JCLI-D-11-00650.1, 2012.

Kerminen, V. M. and Kulmala, M.: Analytical formulae connecting the real and the apparent nucleation rate and the nuclei number concentration for atmospheric nucleation events, J. Aerosol Sci., 33(4), 609–622, doi:PiiS0021-8502(01)00194-X, 2002.

Liu, X., et al.: Toward a minimal representation of aerosols in climate models: Description and evaluation in the Community Atmosphere Model CAM5. Geoscientific Model Development, 5, 709-739, doi:10.5194/gmd-5-709-2012, 2012.