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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Indirect radiative forcing by ion-mediated nucleation of aerosol" *by* F. Yu et al.

Anonymous Referee #2

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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 lim-

itations 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 2^{nd} 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 2^{nd} indirect aerosol effect (Albrecht, 1989), and to some extent the side effect of dynamics responding the the concurrent change in cloud albedo via the 1^{st} 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 2^{nd} indirect effect Albrecht (1989). I would like to ask the authors to discuss this in 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?

Comparison with observations

The authors have conducted a comparison/evaluation of their results with seleceted 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.

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).

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.

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.

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 $\sim$25-70 nm 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.

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.

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?

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 SO_2 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: SO_2 emission plumes take up a very small volume compared to the grid volumes of climate models. Letting the all anthropogenic sulfur being emitted as SO_2 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 $1.9^\circ \times 2.5^\circ$ used in this study, correct aerosol concentrations from nucleation in subgrid-scale SO_2 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.

P.7/L.1: H_2SO_4 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. H_2SO_4 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 H_2SO_4 column burdens. It would be helpful to add a few sentences that say, e.g., that the high H_2SO_4 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 H_2SO_4, ..."

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?

References

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