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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol nucleation and its role for clouds and Earth's radiative forcing in the aerosol-climate model ECHAM5-HAM" *by* J. Kazil et al.

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We would like to thank the reviewer for the helpful comments which have lead to substantial improvements in the manuscript.

1. There are still large uncertainties in our knowledge of atmospheric nucleation and authors also discuss some of these uncertainties in the introduction. However, I feel that the authors are overconfident about the accuracy of the applied nucleation parameterizations. To my knowledge even neutral sulfuric acid-water nucleation in the laboratory conditions cannot be theoretically modeled with the accuracy of one order of magnitude in nucleation rate (e.g. Brus et al, ACP 10, 2631-2641, 2010).

There is a fundamental difference between our treatment of nucleation and the calculation of the aerosol nucleation rate based on classical nucleation theory, as in Brus et al. (ACP 10, 2631-2641, 2010): Classical nucleation theory determines the Gibbs' free energy of formation of small molecular clusters assuming that these are characterized by the surface tension and vapor pressure of a bulk solution with the same composition as the clusters. This approach breaks down for very small clusters, because the concepts of surface tension and bulk solution do not apply to particles containing only a few molecules. Our neutral/charged sulfuric acid/water nucleation nucleation scheme operates with laboratory-measured thermochemical parameters (enthalpy/entropy change of formation, see manuscript for references) of the small neutral and charged sulfuric acid/water clusters, which imposes a very strong experimental constraint on the resulting nucleation rates.

While the applied theoretical models in the current study are sound on their own right, they are not yet fully verified against laboratory measurements, and such claims should not be made.

We have replaced the two ambiguous sentences

- "Here, we investigate three aerosol nucleation processes that are quantitatively well established by laboratory or field experiments in a global climate model." (Introduction)
- "Three aerosol nucleation mechanisms that are quantitatively well established by laboratory or field measurements were incorporated into the aerosol-climate model ECHAM5-HAM:" (Conclusions)

with

- "Here, we investigate in a global climate model three aerosol nucleation processes that are described based on laboratory or field measurements." (Introduction)

- "Three aerosol nucleation mechanisms, described based on laboratory or field measurements, were incorporated into the aerosol-climate model ECHAM5-HAM:" (Conclusions)

It also remains open how well the applied parameterizations represent real atmospheric nucleation. Therefore, errors of around one order of magnitude in the modeled nucleation rate cannot be considered as "very large", as claimed by authors. I think it should be clearly stated that these kind of errors may arise due to uncertainties in the applied nucleation parameterizations.

We have revised the corresponding passage of manuscript to read as follows:

- "However, nucleation parametrizations may exhibit errors in excess of one order of magnitude due to underlying uncertainties. We have repeated simulation $\mathcal{S}_{\text{ref}}$ with neutral and charged H_2SO_4 /H_2O particle formation rates reduced to 1/10 of their original values."

2. The importance of charged nucleation in the atmosphere is currently debated. Previous global model studies have suggested that neutral nucleation can explain the vertical CN profile reasonably well ...

Neutral nucleation is equally debated: Hanson and Lovejoy (J. Phys. Chem. A, 2006, 10.1021/jp062844w) have shown that laboratory measurements do not support efficient neutral nucleation of sulfuric acid and water at temperatures in the middle and lower troposphere. Other laboratory studies (Berndt et al., Science, 2005, 10.1126/science.1104054; Berndt et al. GRL, 2006, doi:10.1029/2006GL026660) do support an efficient neutral nucleation at temperatures in the middle and lower troposphere.

... while the current study applying both charged and neutral nucleation parameterization overestimates the free tropospheric concentrations.

This is also the case when only neutral nucleation according to Vehkamäki et al. (JGR, 2002, 10.1029/2002JD002184) is used: Zhang et al. (ACP, 2010, doi:10.5194/acp-10-6409-2010, Figure 9) have found that ECHAM5-HAM severely overestimates the observed ultrafine aerosol profiles over the tropical and north Pacific (compared to observations and other models). The reason is, as discussed below, that ECHAM5-HAM is biased towards high ultrafine aerosol concentrations, independently of the nucleation scheme.

I have no doubt that the selected set of nucleation mechanisms is plausible according to our current knowledge. But does the inclusion of charged nucleation improve the modelled CN profile at all? I suggest the authors show the vertical profiles from $\mathcal{S}_{\text{noc}}$ and $\mathcal{S}_{\text{non}}$ simulations (perhaps in Figure 4).

Switching off charged nucleation leads to improved ultrafine aerosol profiles, but does not eliminate the bias. The same holds for switching off neutral nucleation. We have also tested the parametrization of neutral nucleation of Vehkamäki et al. (JGR 2002) using the present version of the model, which produces a much more severe overestimation of the observed ultrafine aerosol concentrations. ECHAM5-HAM therefore appears biased towards high ultrafine aerosol concentrations, independently of the nucleation scheme. We have added the following sentence in the revised version of the manuscript:

- "... switching off charged or neutral nucleation entirely does not eliminate the overestimation in the simulated ultrafine aerosol concentrations compared to the observations, indicating a general bias of the model towards high ultrafine aerosol concentrations that is independent of the nucleation scheme."

Authors should discuss how the overestimation of the free tropospheric CN and marine cloud droplet burden affect the calculated contribution of nucleation to short wave radiation.

We have added the following sentence in the discussion of the uncertainties in the revised manuscript:

- "An overestimation of the fraction of cloud droplets that have formed from aerosol nucleation may result in an overestimation of the contribution of aerosol nucleation to net TOA shortwave radiation."

1. How is the activation nucleation limited to the forested area in the model?

In every model grid volume below the boundary layer height, the number of particles from cluster activation is scaled by the fraction of land covered by forest. The forest fraction is taken from a land use table. We have added this in the revised version of the manuscript.

Does the model take into account the secondary organic concentrations in the implementation of this nucleation mechanism, and the seasonal cycle of organic emissions? Please explain in more detail. Also the secondary organics scheme applied in the model should be explained.

There is no secondary organic aerosol scheme in the ECHAM5-HAM model version used in the present work. The parametrization of cluster activation (Sihto et al., ACP, 2006, 10.5194/acp-6-4079-2006) does not account for concentrations of gas phase organic molecules, only for the concentration of gas phase sulfuric acid. We have added the following sentences in the revised version of the manuscript:

- "With the exception of sulfur compounds, aerosol mass originates from primary emissions in the present version of ECHAM5-HAM. This is a realistic assumption for most of the treated species, but may result in an underestimation of particulate organic matter."

- "This formulation does not represent a possible dependence of cluster activation on the concentration of gas phase organic molecules, and may overestimate the resulting nucleation rate at locations where organic molecules are lower than in the boreal forest, such as in the free troposphere and over the oceans."

2. The authors make a claim that activation nucleation should only be applied for the forested boundary layer only. There is no hard evidence to support this claim.

We write that:

- ... we do not exclude that cluster activation, and more generally nucleation involving organics do proceed over the oceans and in the free troposphere. However, it is a fair assumption that they proceed at much lower rates than observed in the boreal forest.
- ... cluster activation, and more generally nucleation involving organic molecules is better suited to explain aerosol nucleation in the boundary layer, in particular over forests, where organic compounds are abundant. At other locations, other nucleation mechanisms, including neutral and charged nucleation of sulfuric acid may play a more important role.
- ... the use of a nucleation parametrization developed from measurements in the forested boundary layer at locations which may substantially differ in gas phase composition and other parameters such as temperature is disputable.

These statements are plausible as the concentrations of organic molecules, which are needed for cluster activation, are found in much higher concentrations in the forested boundary layer than elsewhere. Therefore, the parametrization of nucleation due to cluster activation of Sihto et al. (ACP, 2006, 10.5194/acp-6-4079-2006), which was developed from measurements in the forested boundary layer in Hyytiälä, may overestimate particle formation rates at locations with low concentrations of organic molecules.

We formulate this now explicitely in the revised version of the manuscript, and we now also write in the conclusions section that we do not exclude that cluster activation, and more generally nucleation involving organics do proceed over the oceans and in the free troposphere.

It is possible that activation or kinetic-type nucleation is important in other environments as well. The role of marine organics in activation or kinetic-type nucleation cannot be ruled out completely, and for example Vaattovaara et al. (ACP 6, 4601-4616, 2006) suggested that marine organics can significantly contribute to marine/coastal nucleation.

Vaattovaara et al. (ACP 6, 4601-4616, 2006) propose that iodine oxides are mainly responsible for the observed nucleation; they write in their conclusions

- Taken together, the nucleation mode measurements presented in this paper suggest that iodine oxides are mainly responsible for the nucleation and early growth of newly formed coastal particles, and that condensable secondary organics start more effectively to contribute to the growth at particle sizes of about 5-6 nm.

3. The authors discuss that the overestimation of free tropospheric CN can be caused by the underestimation of condensation/coagulation sinks. I think it would be good to show the resulting CN profile from $\mathcal{S}_0$ (all nucleation disabled) simulation. How much do the primary particles contribute to free tropospheric CN concentrations, and possibly to sinks?

This is indeed a very good point which we realize needs to be better addressed in the manuscript. We see from the $\mathcal{S}_0$ simulation (no nucleation at all) that the contribution of primary aerosol to the aerosol H_2SO_4 condensation sink is extremely small above 4 km over the three considered regions of the Pacific. The reason is that cloud processes in (deep) convection are extremely efficient in removing sea salt particles, which are the

predominant primary aerosol over the Pacific. A very similar result is obtained in the geo-engineering study by Korhonen et al. (ACP, 10, 4133-4143, 2010), who show that injection of artificial sea spray aerosol into the boundary layer has no effect on aerosol number or H_2SO_4 concentration above 4 km. This means that the overestimation of the ultrafine aerosol concentration seen in our results is unlikely due to an underestimation of loss of H_2SO_4 and of freshly nucleated particles onto pre-existing primary aerosol. We have included this important point and the corresponding figure in the revised manuscript.

4. There are not so many global modeling studies about the importance of nucleation as a source of new atmospheric particles, and the ones out there should be mentioned. Particularly Merikanto et al. (ACP, 9, 8601-8616, 2009), Merikanto et al. (ACP, 10, 695-705, 2010), Spracklen et al. (ACP 10, 4775-4793, 2010), Spracklen et al. (GRL 35, L06808, 2008), Pierce and Adams (9, 1339-1356, 2009) and Yu and Luo (ACP, 9, 7691-7710, 2009) and their connection to present work should be discussed.

Done.

5. It would be good to give a value for the overall aerosol forcing given by the model, if authors have calculated such a value. This would be a good reference value when discussing the overall importance of nucleation.

Within the framework of the model we can calculate the radiative forcing of the Earth by a process, which is the contribution of the process to net top-of-the-atmosphere radiation, and anthropogenic forcing via a process, which is the contribution of the process to the change in net top-of-the-atmosphere radiation from pre-industrial to present day conditions. Both can be split up in a long- and a short-wave contribution. In this manuscript, we focus on the short-wave radiative forcing of the Earth due to aerosol nucleation. The radiative forcing by the direct aerosol effect (contribution of aerosol to net top-of-the-atmosphere radiation via the direct effect) amounts to -1.92 W/m^2 (short-wave), 0.17 W/m^2 (long-wave) and -1.74 W/m^2 (total) in the year 2000 global mean of simulation $\mathcal{S}_{\text{ref}}$. The radiative forcing values in the manuscript do not reflect the definition of radiative forcing used by the IPCC, which would require running simulations for pre-industrial and present day conditions.

6. Page 12281, sentence stating "Clouds with fewer but smaller cloud droplets, which formed at higher aerosol concentrations, have a higher albedo and reflect more incoming solar radiation into space at a fixed liquid water path" is incorrect. Higher aerosol concentrations lead to higher droplet concentrations.

This is correct. "fewer" was replaced with "more" in the revised version of the manuscript.

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