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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.

Anonymous Referee #2

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The manuscript by J. Kazil et al. presents the first estimates of the importance of different atmospheric nucleation mechanism on Earth's radiative budget. Using new schemes for neutral and charged nucleation of sulfuric acid and water and activation nucleation of sulfuric acid and an organic compound, the authors estimate the contribution of each of these proposed mechanisms to overall aerosol forcing. The manuscript is well written and includes a thorough analysis of the possible sources of error in the implementation of the new nucleation schemes in ECHAM5-HAM. Comparisons between modeled and measured CN concentrations above ocean regions form the backbone of model validation. The CN measurements by Clarke and Kapustin represent a good reference dataset for general model validation of the total aerosol number

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Interactive Discussion
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in ocean regions, but tell little how the model performs over continental sites. However, together with comparisons between measured and modeled cloud droplet burdens the presented comparisons between model results and measurements are sufficient for the scope of the paper.

Overall, this work is relevant for our understanding of impact of nucleation on climate, and in the scope of ACP. However, a the following comments should be addressed before the manuscript can be published in ACP:

Major comments:

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). 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. 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.

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

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elled CN profile at all? I suggest the authors show the vertical profiles from S_{noc} and S_{non} simulations (perhaps in Figure 4). 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.

Minor comments:

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

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. 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.

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 S_0 (no nucleation at all) simulation. How much do the primary particles contribute to free tropospheric CN concentrations, and possibly to sinks?

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,

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7691-7710, 2009) and their connection to present work should be discussed.

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.

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 12261, 2010.