

## ***Interactive comment on “Atmospheric nucleation: highlights of the EUCAARI project and future directions” by V.-M. Kerminen et al.***

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We would like to thank the reviewer for his/her positive comments and suggestions for improvements. Our detailed response to each of these comments is given below.

Comment: 1) Page 16500, Line 12: Please also cite “Kazil, J., Stier, P., Zhang, K., Quaas, J., Kinne, S., O’Donnell, D., Rast, S., Esch, M., Ferrachat, S., Lohmann, U., and Feichter, J.: Aerosol nucleation and its role for clouds and Earth’s radiative forcing in the aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys. Discuss., 10, 12261-12308, doi:10.5194/acpd-10-12261-2010, 2010.”, and “Luo, G., and F. Yu, A numerical evaluation of global oceanic emissions of alpha-pinene and isoprene, Atmos. Chem. Phys., 10, 2007-2015, 2010.”

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Interactive Discussion

Discussion Paper



We added the paper by Kazil et al. (2010) into the text. The paper by Luo and Yu (2010) deals with oceanic emissions of organic compounds and does not fit into this context.

Comment: 2)Page 16503, Eqn. 1: The coagulation sink is a strong function of size. Why use the coagulation sink of 2 nm particles rather than something in between 2 and 3 nm? The difference in CS between 2 nm and 3 nm is close to a factor of 2.

We agree with the reviewer. The problem, however, is that some sort of approximation is needed due to lack of data (particle number size distribution between 2 and 3 nm). The text was modified as follows: “Since the particle number size distribution in the size range 2-3 nm is not known, we cannot calculate the exact rate at which particles in this size range are lost by coagulation with pre-existing larger particles. As a result, we use CoagS2 as an approximation to this loss rate, which may lead to a slight over-prediction of the value of  $J_2$ .”

Comment: 3)Page 16506, Line 28 through page 16507, line 7: Because ion-induced nucleation cannot be detected when growth rates are slower (please provide a rough cutoff growth rate for when IIN can no longer be detected), won't this bias this technique towards finding the ion-induced nucleation is less important than it is? IIN can only be found if the growth rates are also fast.

The framework by Kerminen et al. (2007) referred to in the paper helps one to identify the days when particles grow too slowly for the analysis of the fraction of ion-induced nucleation, and which should therefore be left out from the analysis. This holds for all the nucleation event days when the particles grow too slowly, including those when particles may have formed solely by neutral pathways. If this is taken into account, no bias toward too low ion-induced fraction should results (As a matter of fact, theoretical studies indicate that charged cluster may grow more rapidly than neutral ones, so the bias should be toward larger rather than lower ion-induced fractions.

Comment: 4)Page 16521, line 24: Please cite Yu, F., Ion-mediated nucleation in the

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atmosphere: Key controlling parameters, implications, and look-up table, *J. Geophys. Res.*, 115, D03206, doi:10.1029/2009JD012630, 2010.

The reference was added.

Comment: 5)Page 16523, top paragraph: How sensitive are the inferred organic condensation rates to the uncertainties in the H<sub>2</sub>SO<sub>4</sub> condensation (either from CIMS or proxy) and the concentrations and growth rates of 2-4 nm particles? Has this sensitivity test been done?

The error in the organic condensation rate is directly proportional to the error in the “unexplained” fraction of the growth rate of 2-4 nm particles. Besides uncertainties in H<sub>2</sub>SO<sub>4</sub> concentration, the error in this fraction is affected by the accuracy by which the growth rate itself can be determined from the measurement data. The error sources are now explained in more detail in a new paragraph added on page 16523 (after line 20, see also the comment 2 by the other reviewer):

“There are two issues worth mentioning here. First of all, the derivation of semi-empirical parameterizations, like Eqs. 4-11, is always subject to uncertainties in measured quantities. For example, there is up to 50% uncertainty in measured H<sub>2</sub>SO<sub>4</sub> concentrations and even a slightly larger one in [NucOrg] due to uncertainties related to determining the growth rate of 2-4 nm particles (Paasonen et al., 2010). Likewise, formation rates of 2-nm particles ( $J_2$ ) may be up to a factor two lower or higher than the estimated ones due to uncertainties in measurements and data analysis (Manninen et al., 2010). Second, it is clear that Eqs. 4-11 are oversimplifications of the physical and chemical factors influencing the nucleation rate. These include the ambient temperature and relative humidity and the stabilizing effect of vapors other than H<sub>2</sub>SO<sub>4</sub> and NucOrg, causing additional scatter in measured data points. Such factors need to be investigated more thoroughly in the future, along with the applicability of the current parameterizations for conditions other than continental boundary layers.”

Comment: 6)Page 16523, lines 9-20: Please quantify the goodness of each parame-

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terization. Give correlation coefficients or a table.

The goodness (correlations, variability etc.) of these parameterizations have been studied in detail by Paasonen et al. (2010), and it is not intended to repeat that thorough description here. The main results are anyway summarized on page 16523 (lines 9–19). Furthermore, we added a new paragraph into the revised manuscript to discuss various uncertainties related to these parameterizations (see the comment above).

Comment: 7)Page 16524, line 16: Pierce and Adams 2007 not in references.

The reference was added to the reference list.

Comment: 8)Page 16525, line 9: Why not show the Anttila (2010) growth parameterization here instead of or as well as the Lehtinen (2007) one?

We were a bit unclear in the original manuscript. Formally, the parameterization by Anttila et al. (2010) is the same as Lehtinen et al. (2007) (equation 12), but it has an iterative procedure to take into account the effect of self-coagulation. We modified the paragraph into the following form:

“A drawback of equation (12), like in all other corresponding parameterisations developed until now, is the neglect of nuclei self-coagulation. This process accelerates nuclei growth and reduces their number concentration. Anttila et al. (2010) derived an iterative procedure by which the effect of nuclei self-coagulation on GR and CoagS can be taken into account when applying equation (12). Comparisons to detailed numerical simulations showed that the apparent particle formation rate is affected by nuclei self-coagulation only when atmospheric nucleation rates are exceptionally high ( $>10 \text{ cm}^{-3} \text{ s}^{-1}$  in the free troposphere and  $>104 \text{ cm}^{-3} \text{ s}^{-1}$  in the polluted boundary layer).”

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 16497, 2010.

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