

Interactive comment on “Ion-mediated nucleation as an important global source of tropospheric aerosols” by F. Yu et al.

Anonymous Referee #4

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The paper deals with a very important subject of modelling the contribution on ion-mediated nucleation to atmospheric nucleation rates. The subject is still highly debatable in many aspects and therefore new insights on the phenomena are needed. However, in my opinion the MS is not publishable in its' current form for the reasons given below:

1) As also the authors note, the role of ion induced nucleation and even the possible ion-induced nucleation mechanisms are somewhat unresolved. Taking this into account, the authors treat the subject from a slightly too narrow and purpose-oriented point of view. Because of these clear discrepancies between different studies, the authors should very carefully demonstrate e.g. their model calculations and the uncertainties in their approach. The authors for instance imply that e.g. Laakso et al.

(2007) have interpreted their data on atmospheric charged fraction in an incorrect way. When making this kind of statements, the authors should explicitly give the concrete physical aspects of their approach that differ from the data interpretation of Laakso et al. (2007). Now the differences between the two approaches are not clear.

2) Besides the afore-mentioned work by Laakso et al. (2007) also recent studies by e.g. Iida et al. (2006) and Kulmala et al. (2007) based on atmospheric measurement data from the boundary layer indicate that the observed ion concentrations and charged fractions are not enough to explain the observed total particle formation rates in the considered sites (Boulder, Colorado, US and Hyytiälä, Finland). What do the authors think about these studies? These studies should be also commented more thoroughly in the paper to reflect the different scientific conceptions of the role of ion-mediated nucleation.

3) In my opinion it is not enough to include only results on the ion-induced/ion-mediated nucleation in this kind of paper. Similar calculations should be presented also for other nucleation mechanisms (e.g. binary and ternary nucleation, or the semi-empirical cluster activation theory as done by e.g. Spracklen et al., 2006), at least for some sites. Without this kind of comparison it is impossible to assess the relative role of ion-induced nucleation. Therefore I think that the MS is not stand-alone enough to be published as an independent paper. Particularly statements like "The general agreement between simulations and observations demonstrated above strongly supports the important role of IMN in generating new particles in global troposphere" (p. 13609) should not be made without demonstrating how the other approaches succeed in producing qualitatively correct results.

4) I agree with Ari Laaksonen's interactive comment that the authors should present a detailed exemplary analysis for at least one site where they demonstrate the model calculations. For instance an exemplary day with measured values of SO₂ and sulphuric acid concentrations, ion production rates, particle and ion size distributions, radiation, temperature and atmospheric particle growth and formation rates are used should be

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presented.

5) Related to the previous comment, the authors should comment on how the dynamics of the aerosol size distributions and the gas concentrations are treated in the model. It has been observed in several studies that e.g. the boundary layer dynamics as well as the daily evolution of the aerosol size distribution (the evolution of e.g. the CS and the nucleation mode) affect strongly the occurrence of nucleation and behaviour of the freshly formed nucleation mode. Does the nucleated mode contribute to the values of the CS?

6) The model input values for the different sites could be presented in similar look-up tables that has been done for the experimental data.

References:

Iida, K. et al., J. Geophys. Res., 111, D23201, doi:10.1029/2006JD007167, 2006.

Kulmala, M. et al., Science, 318, 89, 2007.

Laakso, L. et al., Atmos. Chem. Phys., 7, 1333, 2007.

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