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Interactive comment on “Using measurements of the aerosol charging state in determination of the particle growth rate and the proportion of ion-induced nucleation” by J. Leppä et al.

Anonymous Referee #3

Received and published: 19 October 2012

In this manuscript the authors tested the applicability of several data analysis methods to determine the growth rate and the proportion of ion-induced nucleation from the measured charged fractions. The approach is to compare the growth rate and initial fraction of charged particles estimated from these methods with the values obtained directly from the aerosol dynamic simulations. The authors found that the accuracy of the data analysis methods depends on a number of factors, and concluded that the existing data analysis methods should not be used when the nuclei growth rate is less than $\sim 3\text{nm/h}$, or when charged particles grow much more rapidly than neutral ones. Measured charged fractions of freshly nucleated particles, if properly interpreted, can provide very useful insights about the mechanisms of new particle formation. The

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analysis and comparison presented in this study is useful to understand the applicability and uncertainty of the simplified data analysis methods. The following comments should be properly addressed before the publication of the manuscript in ACP.

1. In the last couple of years, quite different conclusions have been derived from the same measured charged fractions about the relative importance of ion-induced or mediated versus neutral nucleation processes: one based on simplified data analysis method (Laakso et al., 2007; Manninen et al., 2009; Gagné et al., 2010) and the other based on kinetic aerosol dynamic model (Yu and Turco, 2008, 2011). Does the study reported in this manuscript help to reconcile the difference?
2. The simplified data analysis methods have been used to estimate the contribution of ion-induced or ion-mediated nucleation to new particle formation by the authors in a number of previous publications. Based on the new insights obtained in this study about the applicability and uncertainty of the simplified data analysis methods, please discuss the uncertainties in your previous estimation with regard to the contribution of ion nucleation to total particle formation. As you concluded, the method should not be used if growth rate is less than 3 nm/h. According to Manninen et al. (2009), the median GR values for 1.3-3 nm “intermediate” ions at Hyytiälä in spring 2007 were estimated using ion mobility spectra to be \sim 1.9 nm/hr. Does this imply that you can't apply the simplified data analysis methods to estimate the contribution of ion nucleation in a large fraction of nucleation event days observed at Hyytiälä?
3. Yu and Turco (2011) have discussed in detail the evolution of charged fractions of particles of different sizes based on a detailed size-resolved kinetic aerosol dynamic model. I am surprised that Yu and Turco's work is not mentioned at all. It will be useful to compare the results of this study with those reported in Yu and Turco's work.
4. This study uses results from aerosol dynamic simulations as references to assess the applicability of the simplified data analysis methods. What is the uncertainty of aerosol dynamic simulations? How much the uncertainty may affect the conclusions of

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this manuscript?

5. P21869, Lines 11-20. What about ion-mediated nucleation (IMN) which includes IIN as well as the growth of neutral clusters formed by ion-ion recombination but are smaller than critical sizes?

6. P21869, Lines 19-25. Please give the values of the IIN fraction reported in this study. I agree that “The contribution of IIN to new particle formation is important from climate change point of view”. In this regard, the different conclusions about the contribution of IIN or IMN to new particle formation derived from same set of observations (see comment 1 above) are highly relevant and should be discussed here.

7. P21876, Lines 9-11. What are the sizes of small ions assumed in your model? How do you calculate the recombination coefficient of small ions with charged particles?

8. P21886, Lines 14-21. Again the strong dependence of charged fractions on particle sizes has been illustrated in detail in Yu and Turco (2011). Are your results here consistent with those found in Yu and Turco?

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ACPD

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