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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Nucleation mode growth rates in Hyytiälä during 2003–2009: variation with particle size, season, data analysis method and ambient conditions" *by* T. Yli-Juuti et al.

T. Yli-Juuti et al.

taina.yli-juuti@helsinki.fi

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Response to Dr. K. Iida (referee)

We thank Dr. Iida for his useful and constructive comments. We have now carefully considered them and will take them into account in the revised version of our manuscript. Our detailed answers to Dr. Iida's comments are listed below:

Specific Comments

Page 21277 Equation 6. The value of k_{H2SO4} is not given.

We used k_{H2SO4} determined by Petäjä et al. 2009: $k_{H2SO4} = 8.4e-7 * UVB^{-0.68}$ (m^2 W^{-1} s^{-1}). This information will be added in the revised manuscript.

Page 21280 Line 5. "On the other hand, the method is not very sensitive to size dependent process..." This statement does not seem consistent with what stated in line 16-20. In line 16-20 authors state that scavenging, which is size dependent, tends to overestimate GR obtained by this method.

We agree with Dr. Iida that this was somewhat unclearly formulated in the manuscript. What we mean is that rather than any size-dependent coagulation losses per se, it is the change in time of the sink caused by coagulational scavenging, rather than the existence of this size dependent sink, which affects the GR calculated with maximum concentration method. We will clarify this in the revised manuscript.

Page 21280, Line 19-21 "this GR calculation may fail as the moments of maximum concentration would be shifted in time due to particle loss..." It is recommended that authors provide a bit more information so that readers can understand the authors' arguments. How about something like this? "CS generally increases with time after nucleation starts. Younger particles would be depleted more than older particles when they reach the same size, which tends to distort the profile of concentration vs time to earlier time." In addition, concentration of condensing vapor (therefore GR) can also change with time during NPF period. Authors can qualitatively discuss how the time dependence in true GR can alter the GRs estimated by the maximum concentration method.

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These are good points and we will clarify these (and add discussion on the time-dependence of the condensable vapour concentration) to the revised manuscript. We will also add a reference to the manuscript by Kuang et al. (ACPD 2011) that discusses these issues – namely the size- vs. time-dependence of the growth rates.

Kuang, C., Chen, M., Zhao, J., Smith, J., McMurry, P. H., and Wang, J.: First size-dependent growth rate measurements of 1 to 5nm freshly formed atmospheric nuclei, Atmos. Chem. Phys. Discuss., 11, 25427–25471, 2011. www.atmos-chem-phys-discuss.net/11/25427/2011/

Page 21282, Line 24-26 "if the condensation sink stays constant in time...." Including the effects of scavenging and self-coagulation as a part of maximum-concentration method seems to require similar calculations done in mode-fitting method. The maximum concentration method does not seem to be more advantageous than mode-fitting method when one try to account for scavenging and self-coagulation.

As we see it, the impacts of the coagulation scavenging and self-coagulation on the mode-fitting method and the maximum-concentration method are slightly different. While the mode-fitting method is affected by the "coagulational narrowing" of the size distribution at each moment in time – due to the size-dependence of the coagulational scavenging rate, the impact on the maximum concentration method is mainly through the time-dependence of the coagulation sink. It would, in principle be possible to correct also the concentrations in the maximum concentration method with the time-dependence of the coagulational scavenging rate, but this correction is not exactly the same as for the mode-fitting method. We will add discussion clarifying these points to the revised version of the manuscript.

Page 21283, Line 1-3 It is clear that maximum concentration method is much more useful if dN/dlogDp vs Dp at given time measured by BSMA or AIS are limited in rel-

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atively narrow size range therefore no distinctive peak can be seen in the measured particle size distributions (PSD). However, if dN/dlogDp vs Dp at given time measured by DMPS data show a distinctive PSD mode shifting with time in >5nm range the mod-efitting method seems more reliable since the procedure to account for coagulation and scavenging are already well-established by Stlozenburg (2005).

These are good points, and it is true that the methods developed by Stolzenburg et al. (2005) are a standard way of correcting for the coagulational losses. However, we would also like to refer to our last response for the clarification on the impact of the coagulation sink to the maximum concentration method: this impact is likely to be important only if there is a significant change in time in the coagulation sink over the time period of when the maximum concentration in a specific size channel is reached – in this case it is possible that the maximum concentration is somewhat shifted in time as compared with the case when only condensational growth would take place. We will add more discussion of these matters.

Page 21285, Line 22-25. It is recommended that authors emphasize somewhere in the text that "different instruments" means not only different instruments (i.e., AIS, BSMA, DMPS) but also operating under different polarity modes.

This will be done in the revised manuscript, as suggested by the referee.

Page 21286, line 8-10 Author can comment whether these differences are potentially caused by the inaccuracies in the calibration of voltage-mobility relation of AIS or not.

We are fairly confident that the differences are not caused by the inaccuracies in the calibration of voltage-mobility relation of AIS. The AIS detects mobilities very accurately as reported by Asmi et al. (2009 ACP). They observed only 0.1 nm inaccuracy in mobility diameters. Therefore, this is not the answer to the difference in GRs determined from AIS and BSMA size distributions. On the other hand, the AISs detect the mobil-

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ities accurately for monomobile ions (Asmi et al. 2009 ACP, Gagne et al. 2011 AMT), while for polydisperse particles the distribution might be slightly skewed.

On average Gagné et al. (2011 AMT) observed higher growth rates with AISs compared to BSMA. They observed some tens of percents variation in GRs between the instruments. This is in agreement with our observation.

The small deviation in GR found here was on the same order of magnitude as was obtained in recent calibrations by Gagne et al. (2011) using instruments with precisely determined flow rates.

Page 21286, Title 3.3.2 should be changed from "the effect of electrical charge" not just "charge".

This will be done in the revised manuscript.

Page 21287, Line 20-26. The relation between median difference and relative difference is not clear since a definition of relative difference in this context is not clearly stated. It is recommended that authors graphically show and compare the distributions of relative standard deviations among different measurement techniques and different GR calculation methods. I believe that it is one of the important conclusions in this paper.

We will clarify the definitions of the uncertainties to the revised version of the manuscript. We agree that the relative difference was possibly misleadingly stated. We have now revised the calculation of relative uncertainty related to methods to be the same as for instrument comparison so that they can be compared more reasonably. This changes the uncertainty related to methods and gives relative uncertainty of 16%, which is comparable to the uncertainty calculated based on different instruments. We will modify the conclusions related to uncertainties accordingly in the re-

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vised manuscript. Please also find below distributions of the relative uncertainties in the whole data set related to a) measurement method and b) analysis method. We would prefer not adding these to the revised manuscript, mostly for the sake of brevity – particularly as they do not show any significantly different implications as compared with the median relative uncertainty.

Page 21288, Line 3-5 There seems to be at least two ways to interpret difference in GR seen in Figure 6a. (1) Particles grew fast below 10nm then slowed down at larger sizes. As a result PSD accumulated above 10nm. If this is true the maximum-concentration method can be more effective way to measure GR. (2) Altitude of air-mass where the nucleation started to occur was greater than the sampling height. The air-mass slowly mixed into the sampling height while particles grew up to 10nm during the mixing. If this is true mode-fitting method is more effective way to measure GR since the peak diameters of PSD are not affected by air-mass-movements. Since the idea of "faster growth rate at smaller sizes" stated in (1) is inconsistent with those observed by AIS or BSMA. Scholars in this field including myself would think (2) sounds more natural therefore inclined toward (2).

We think that these are very good points and will add discussion on the possible role of boundary layer dynamics to the revised version of the manuscript.

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Figures

Figure 1. Distribution of relative uncertainty in GR based on a) comparison of instruments and b) comparison of methods. s_{GR} and mean GR refere to the standard deviation and mean of the GR values determined a) from the data measured with the different instruments or b) with the different GR calculation methods for a NPF event.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 21267, 2011.

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Interactive Comment

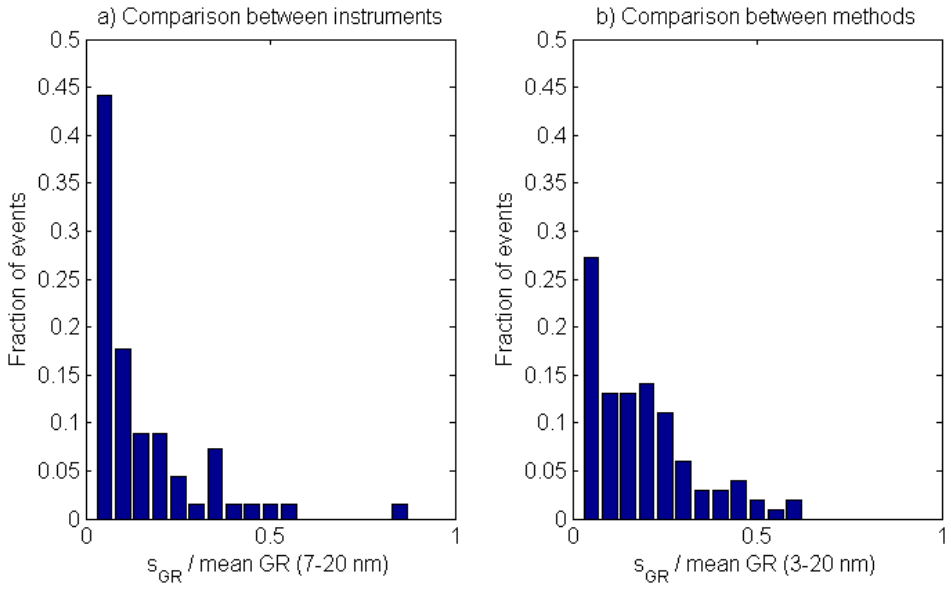

Fig. 1.

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