To: Editor, Atmospheric Chemistry & Physics
From: Manuscript Authors
Regarding: Response to Referees' Comments

We sincerely appreciate the guidance and suggestions provided to us by the editor and referees. Comments from the first and second referees are numbered and labeled with an **R1** and **R2** prefix, respectively. Responses from the authors immediately follow each comment and are indented with a blue font color.

In light of the number of similar comments made by both referees, full responses to those particular comments will be made for referee 1. Responses to referee 2 regarding those similar comments will refer to the corresponding responses to referee 1.

**REFEREE #1:**

**R1.1**

I think the authors should be more careful with the words first and new. Size dependent GRs have been frequently reported in connection to ion spectrometer studies (e.g. Hirsikko et al., 2005; Manninen et al. 2009; Yli-Juuti et al., 2011).

> We acknowledge that the mentioned ion spectrometer studies present important results for size-dependent growth rates obtained over multi-year campaigns allowing for analysis of seasonal variations in nanoparticle growth rates. Citations of the mentioned studies are now included in the introduction.
>
> We wish to clarify and emphasize that our work in this manuscript presents size-resolved growth rate measurements acquired during a relatively short time interval (15 minutes for the NCCN analysis, and 20 minutes for the NCAR analysis) over which it can be reasonably assumed that the concentration of gas-phase species that potentially participate in nanoparticle growth are relatively constant. In contrast with mentioned ion spectrometer studies, our size-resolved growth rate results were obtained from size distribution measurements of the total aerosol (neutral and charged). This de-coupling of the size and time-dependence for ambient growth rates (of the total aerosol) is a new and significant result of this work. The presented data analysis methods allow, for the first time, "the clear interpretation of observed size-dependent growth as a consequence of the particular growth mechanism at work rather than the consequence of time-dependent vapor condensation" (pg. 25430, lines 11 – 14).
>
> In light of the referee's comments regarding the previous work done to measure initial growth rates of nucleated particles, the manuscript title is now changed to:
>
> "Size and time-resolved growth rate measurements of 1 to 5 nm freshly formed atmospheric nuclei."
>
> Also, the last paragraph in the introduction (section 1) is now rewritten to summarize the prior results for size-dependent growth rates and to highlight the

new and novel differences in the methods and growth rate results presented, in comparison to prior work:

"Using size distribution measurements of the total aerosol (neutral and charged) down to ~ 1 nm geometric diameter, novel data analysis methods were developed in this study to de-couple, for the first time, the size and time-dependence of particle growth rates for freshly nucleated aerosol. While earlier studies have presented evidence for size-dependent growth rates of nucleation mode particles, those results were obtained from size distributions of the ambient ion population and were averaged over particle size and growth time (Hirsikko et al., 2005; Manninen et al., 2009; Yli-Juuti et al., 2011). Methods for obtaining size and time-resolved growth rates are presented, along with insights into the processes of nucleation and growth provided by these measurements."

There are also studies estimating the GRs below 3 nm from the time lag between H2SO4 and CPC cut-off size (e.g. Sihto et al., 2006; Metzger et al., 2009; Benson et al., 2011), and they have already noted that sulphuric acid cannot explain all of the early growth (unlike p. 25438, rows 4-6).

We are aware of this method for estimating growth rates and the resulting observations that sulfuric acid condensation accounts for only a fraction of the observed sub 3 nm growth, observations first made by Weber et al. (1997). The mentioned references to sub-3 nm growth rate results in ambient (Sihto et al., 2006) and laboratory environments (Metzger et al., 2010) are now included in section 3.2.

The studies mentioned by the referee present *ambient* and *laboratory* growth rates for sub 3 nm aerosol (based on the time delay between the peaks in $H_2SO_4$ concentration and the concentration of newly formed particles). These growth rates are, by definition, averaged over the time it takes for a particle to grow from ~ 1 nm to 3 nm, which can also often span several hours. Our techniques developed in this manuscript provide, at an "instant" in time, size-resolved growth rates $GR_{OBS}\left(D_p\right)$ down to 1 nm. Size-resolved growth rate enhancements $\Gamma\left(D_p\right)$ can then be calculated from $GR_{OBS}\left(D_p\right)$. To our knowledge, these are the first results for size-dependent enhancements to the observed growth rate down to 1 nm. The enhancements to sub-3 nm particle growth presented in the studies mentioned by the referee do not present size-resolved growth rate enhancements. The following text is now included in section 3.2 to clarify this difference:

"A number of studies have presented evidence of sulfuric acid limited condensation accounting for only a fraction of the observed sub 3 nm growth in ambient (Weber et al., 1997; Fiedler et al., 2005; Sihto et al., 2006) and laboratory experiments (Metzger et al., 2010). Due to the nature of the methods used to obtain sub 3 nm growth rates in those studies, the reported growth rate enhancements are, by definition, averages over the size and time it takes for a

nucleated particle to grow to ~ 3 nm. The growth rate enhancement values presented in Fig. 2a and 2b are the first reported results of size-resolved Γ down to ~ 1 nm geometric diameter, providing a direct indication that species other than sulfuric acid can play a significant role in particle growth below 2 nm geometric diameter."

Also the size distribution of total aerosol from _1.3-5 nm (in mobility diameter = 1-5 nm in geom. diameter) during nucleation events have been presented previously by Sipilä et al., 2009; Lehtipalo et al., 2009; 2010 (p. 25430, rows 25-28).

In our work, freshly nucleated particles are sized according to their electrical mobility diameter, which is fundamentally tied to the physical, or geometric diameter of the particle (Tammet, 1995; Larriba et al., 2011). The referee has mentioned studies that present size distributions of the total aerosol from 1.5 – 3 nm during nucleation events that utilize an aerosol sizing technique based on inversion of the CPC pulse height distributions (Marti et al., 1996; Saros et al., 1996; Weber et al., 1998; Dick et al., 2000), where the sampled particle size distribution is inferred from the grown particle size distribution. This technique is also known as "activation-sizing". To acknowledge the earlier efforts that were made in measuring sub 3 nm size distributions during nucleation events, and to distinguish our electrical mobility based measurements from earlier activation-sizing measurements, the following text is now added in the introduction (section 1):

"While earlier studies have presented sub 3 nm size distributions acquired during nucleation events using activation-sizing techniques (Sipilä et al., 2009; Lehtipalo et al., 2011), mobility-classified size distributions of freshly nucleated aerosol were measured in this study using a DEG UCPC to extend SMPS measurements down to ~ 1 nm (Jiang et al., 2011a)."

I strongly disagree that your results are any more "direct" than GRs calculated from the size distribution using a different method, which you sweep under the carpet as "estimates" or "inferences" (p. 25430 rows 21-24 and p. 25435 rows 12-24). I suggest changing the title, or at least taking away word first, and giving proper credit to previous work on the subject.

The manuscript title has now been changed and the appropriate references have now been included. Any reference to our growth rate results being more "direct" when compared to earlier growth rate results has now been removed, along with references to earlier growth rate results being "inferences". The following text has been amended and added to highlight the differences between our work and earlier work:

"While those results were obtained from size distributions of the ambient ion population rather than from the total aerosol population (neutral and charged),

such as reported here, their reported growth rate size-dependence is largely substantiated by our results in this study."

**R1.2**

I cannot see any good reason to use geometric diameter of particles. As the DEGSMPS system is measuring particle mobility, I would recommend sticking to mobility diameter to get rid of a redundant conversion and assumptions about particle density (which is not known). This would also make it easier to compare to other papers reporting GRs and measurements of small particles, which usually give the size in mobility diameters. Please change this in text and title, and in figures put mobility diameter to lower abscissa.

The determination of particle growth rates is fundamentally a determination of the rate of particle volume change per unit time. A measure of the particle volume is the particle geometric diameter (Tammet, 1995). Use of the particle geometric diameter when characterizing sub 3 nm particles is therefore physically justified, and indeed necessary. Also, it is not necessary to know particle density to convert between mobility and geometric size. For the sake of clarity and consistency, all references to "mass diameter" in the manuscript have now been removed and replaced with "geometric diameter."

To facilitate comparison with growth rates reported in other studies, we have reported growth rates as functions of both geometric diameter and mobility diameter, since conventional particle sizing is accomplished through particle mobility classification. The relationship between geometric and mobility particle diameters has been established and studied theoretically and experimentally (Tammet, 1995; Larriba et al., 2011). Conversion from mobility to geometric diameter (in nanometers) is simple and straightforward to apply: $D_p^{mob} = D_p^{geo} + 0.3$. We firmly believe that the relationship between mobility and geometric diameter, firmly established by Larriba et al. (2011), is pertinent for the size range analyzed.

The size ranges in figures should also be consistent (C2 and C3a have different axis than all others, and Figure A4 even uses mass diameters).

The y-axis ranges in the figures mentioned by the referee are meant to make the dynamic range in the presented quantities more easily seen and examined. For consistency, the abscissa axis label in Figure A4 has been changed to "Geometric Diameter."

**R1.3**

Your method to calculate the GR is strongly dependent on the shape of the particle size distribution. The results by Jiang et al. (2011) describing the performance of the DEG-SMPS show that the detection efficiency is highly sensitive to particle composition in smallest sizes. As you mentioned, the charging efficiency of smallest particles is not well known, and it might also be dependent on particle composition. How would

the sensitivities to composition affect the measured size distribution and thus GR? (Composition might change with particle size, and also in time).

It is true that the particle growth rates obtained with these population balance methods are influenced by the particle charging and detection efficiency. The effect of reasonable uncertainties in both efficiencies (charging and detection) on the growth rate enhancement Γ has already been presented in Figure C3 and discussed in the manuscript appendices (Appendix C2). The relative effect of these same uncertainties on the growth rate (GR) is identical to the effect on Γ since GR scales linearly with Γ according to Eq. (A7) in the appendices:

$$GR = \Gamma \cdot GR_{SA} \qquad \text{(A7)}$$

where $GR_{SA}$ is the growth rate based on the condensation of only $H_2SO_4$ vapor. The effect of systematic uncertainties in particle charging and detection efficiencies on the growth rate GR (for the NCAR NPF event) as a function of particle mobility diameter and geometric diameter is shown below in Figure R1.3.

In response to the referee's comments, the following text has been added in Appendix C2.1:

"The relative effect of these same uncertainties on $GR$ is identical to the effect on Γ since $GR$ scales linearly with Γ according to Eq. A7 ($GR = \Gamma \cdot GR_{SA}$) in the appendices, where $GR_{SA}$ is the growth rate based on the condensation of only $H_2SO_4$ vapor."

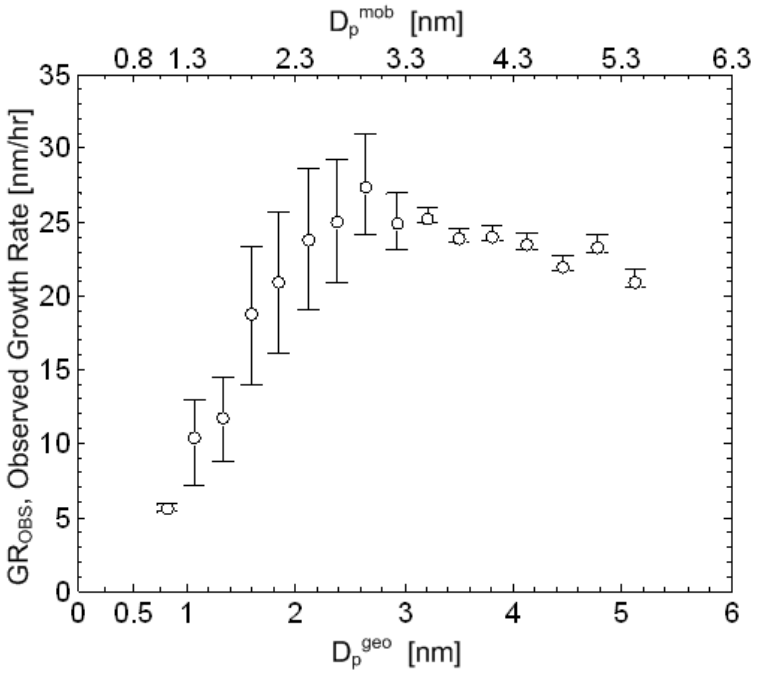

Figure R1.3: Effect of systematic uncertainties in particle detection and charging efficiencies on growth rate $GR_{OBS}$ as a function of particle mobility diameter $D_p^{mob}$ (upper abscissa) and particle geometric diameter $D_p^{geo}$ (lower abscissa) for the NPF event observed on 19 September 2010 (13:00, NCAR).

As the total detection efficiency is very low especially in sizes below 1.5 nm (please give a number also in the main text, and include the SMPS transmission efficiency to figure C2, as in equation A19), the raw counts need to be multiplied with a very large number. What is the range of actual raw counts in the smallest channels that are used to calculate the size distribution? A lot of effort has clearly been put into getting an error estimation for the GRs, however, the estimate does not make any sense if you ignore some of the biggest sources of error. I think these issues should be discussed in more detail also in the main text and not only in the appendix.

As pointed out by the referee, the total detection efficiency for sub 2 nm particles is quite low, and is dominated overwhelmingly by the particle charging efficiency in the neutralizer and the particle detection efficiency in the DEG-UCPC (which also happen to be the quantities that are dependent on particle composition). Values and reasonable ranges for the product of those efficiencies ( $f_{\text{detection}} \cdot f_{\text{charging}}$ ) are already discussed in Appendix C2 and presented in Figure C2.

In terms of the total detection efficiency (from sampling inlet to CPC optics block), the product $f_{\text{detection}} \cdot f_{\text{charging}}$ does, in fact, represent the largest source of uncertainty due to uncertainties in particle composition and charging efficiency below 3 nm. Uncertainties in the total detection efficiency due to sampling losses are a function of uncertainties in the sample flow rates, which were calibrated twice daily and were accurate to within 5%, as discussed in Appendix 2.3.1. For the NCAR NPF event, the average number of counts in the 4 smallest size bins (1.1, 1.3, 1.6, and 1.9 nm) during the period of peak particle production (20 minutes) were 3, 4, 7, and 15 particle counts. As mentioned in the appendices, the uncertainties associated with these raw counts were calculated and fully propagated in the calculation of the size distribution and subsequent determination of the growth rate.

The effects of reasonable systematic uncertainties in $f_{\text{detection}} \cdot f_{\text{charging}}$ on both $\Gamma$ and GR are both qualitatively and quantitatively minor, where trends in both $\Gamma$ and GR as functions of particle diameter are preserved, as evidenced in Figure R1.3 and Figure C3. Presentation of this discussion in the appendices is appropriate since it does not substantively change the main results presented in the main text of the manuscript.

**R1.4.** Based on reading only the main article it is impossible to understand the method for calculating the GR, and especially the difference between the regional and plume event (I think they are more like variations of the same method than two completely

new methods as stated e.g. in p. 25433 row 9). I suggest including at least equations A1 and A16 from appendix to chapter 2.2. together with a bit more description of the method and discussion about the assumptions that are needed for using it.

A concise summary of the data analysis methods involved in obtaining growth rates from the measured size distribution would be helpful to the reader. The following text has been added in section 2.2:

"Size and time-dependent observed particle growth rates, $GR_{OBS}$, were estimated using two new methods based on fitting measured size distributions to the aerosol general dynamic equation (GDE) (Gelbard and Seinfeld, 1978). Whether the measured size distributions were consistent with sampling from a regional air mass or with interception of a local plume determined the appropriate method to obtain growth rates from the measured size distributions. The analysis method for plume events utilizes the novel result that size distributions ($< \sim 5$ nm) for a nucleating system in the presence of an aerosol achieve pseudo steady-state shortly after the start of nucleation (McMurry, 1983). For regional events, the analysis method is similar in principle to earlier analysis techniques (Lehtinen et al., 2004; Verheggen and Mozurkewich, 2006) that fit size distributions to the GDE. For an aerosol system that is growing through simultaneous gas uptake and coagulation, the aerosol GDE can be integrated to describe the evolution of the number concentration between particle diameters $D_{p1}$ and $D_{p2}$ ($D_{p2} > D_{p1}$) according to (Eq. 1):

$$\frac{dN_{\Delta}(t)}{dt} = n(D_{p1},t)\cdot GR(D_{p1},t) - n(D_{p2},t)\cdot GR(D_{p2},t) + CoagSrc_{\Delta}(D_{p1},D_{p2},t) - CoagSnk_{\Delta}\left(D_{p1},D_{p2},t\right) \quad (1)$$

where $\Delta$ is the size interval defined by $D_{p1}$ and $D_{p2}$, $GR = dD_p/dt$, and $n = dN/dD_p$. In the RHS of Eq. (1), the first and second terms are the condensational flux into and out of the aerosol size interval defined by $\Delta$, $CoagSrc_{\Delta}$ is the source term defining the production of particles in $\Delta$ due to coagulation, and $CoagSnk_{\Delta}$ is the sink term defining the removal of particles in $\Delta$ due to coagulation. With a measured size distribution $n$, the only unknown quantities in Eq. (1) are the diameter growth rates at the interval boundaries, $GR(D_{p1},t)$ and $GR(D_{p2},t)$, which are then obtained as functions of time and particle diameter through an iterative solution of Eq. (1) at various particle sizes. Further details of each method can be found in Appendices A1 (regional event analysis) and A2 (plume event analysis)."

**R1.5.** How was the time periods in Fig 1 chosen?

In Figure 1a (NCCN NPF event), the time periods presented were chosen to illustrate the time of day dependence for the observed growth rates (morning

versus afternoon). In Figure 1b (NCAR NPF event), the time period presented (12:40 – 13:00) was during the peak of particle production, where there was adequate particle counts in the smallest size channels.

Seeing the evolution of the particle size distribution (maybe as a surface plot) for the given example days might help to understand the particle dynamics better.

Shown below in Figure R1.5 is a contour plot of the aerosol size distribution of the NCAR NPF event on 19 September 2010.

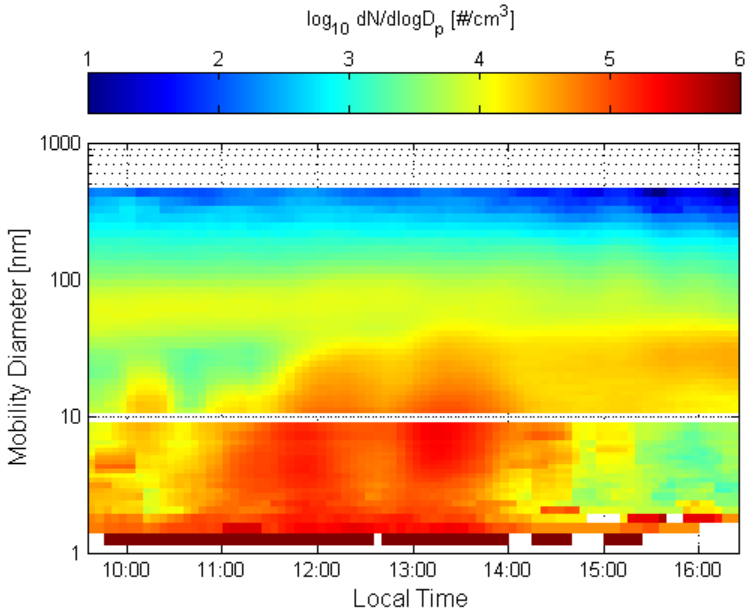


Figure R1.5: Contour plot of aerosol number size distribution during period of particle production and growth as a function of mobility diameter and local time for NCAR NPF event measured on 19 September 2010.

How do the GRs compare to those calculated from the conventional SMPS?

For the NPF events that were analyzed in this study, it was not always possible to compare growth rates obtained with the conventional SMPS (10 – 500 nm) and the DEG-SMPS (1 – 10 nm) for several reasons. For the NPF events measured in Atlanta during the NCCN campaign, the majority of measured size distributions were indicative of plume interception, where it was not possible to obtain growth rates in the > 10 nm size range through conventional methods (e.g., time shift analysis, modal diameter tracking) because our measurements at a fixed sampling site did not allow us to measure size distributions as they evolved in time. For the NCAR NPF event shown in Figure R1.5 above, a modal diameter growth rate of ~ 5 nm/hour was estimated for the growing mode averaged over the time interval 14:00 – 18:00 and averaged over the size interval 10 – 30 nm. It would be difficult to make a direct comparison between this "averaged" estimate and the

higher growth rate values presented in Figure 1b of the manuscript due to the differences in analysis method, aerosol size range, and time interval.

Is the fit between DEG-SMPS and Cluster-CIMS always as good as in Fig. A4 (in text you state that they are in qualitative agreement)?

We find that, during NPF events measured during NCCN, SMPS and Cluster CIMS data are in good qualitative agreement (Jiang et al., 2011b) if CPC detection efficiencies are assumed equal to values measured for NaCl (Jiang et al., 2011a; Kuang et al., 2012) and charging efficiencies are calculated according to bipolar stationary state theory (Wiedensohler, 1988; Reischl et al., 1996; Alonso et al., 1997). No "fitting factors" were used in order to provide agreement between instrument data which were acquired through independent measurement techniques. There were times prior to and after the nucleation event when the Cluster-CIMS detected signals but the DEG-SMPS did not; at those times, the sulfuric acid concentration was low – see Figure 1, 7:15 (Jiang et al., 2011b). Chamber studies of nucleation driven by $SO_2$ photo-oxidation, not yet published, also show similar agreement between Cluster CIMS and DEG SMPS meaurements.

**R1.6.** In the abstract, I assume that the given GRs and enhancement factors refer to the one example case you discuss in detail. Please state this more clearly - at first I thought that the given GR range is the variation of GRs between different days/times (which would be also interesting to know, maybe even as a figure).

The following text has been amended and added to the abstract to more clearly convey that the presented ranges in growth rates and enhancements correspond to ranges in particle diameter for the two NPF events that were discussed:

"One NPF event from each campaign was analyzed in detail. At a given time during the NPF event, size-resolved growth rates were obtained directly from measured size distributions and were found to increase approximately linearly with particle size from ~ 1 to 3 nm geometric diameter, ranging from 5.5 ± 0.8 to 7.6 ± 0.6 nm $h^{-1}$ in Atlanta (13:00) and from 5.6 ± 2 to 27 ± 5 nm $h^{-1}$ in Boulder (13:00)."

Please give the numbers in the same order (Atlanta before Boulder) as they are usually discussed in the text.

Done.

The sentence about enhancement factors (p. 25428, rows 13-19) is also very long and hard to follow, could you reformulate it?

The text in question has been split, to yield:

"The resulting growth rate enhancement $\Gamma$, defined as the ratio of the observed growth rate to the growth rate due to the condensation of sulfuric acid only, was found to increase approximately linearly with size from ~ 1 to 3 nm geometric diameter. For the presented NPF events, values for $\Gamma$ had lower limits that approached ~ 1 at 1.2 nm geometric diameter in Atlanta and ~ 3 at 0.8 nm geometric diameter in Boulder, and had upper limits that reached 8.3 at 4.1 nm geometric diameter in Atlanta and 25 at 2.7 nm geometric diameter in Boulder."

**REFEREE #2:**

**R2.1** The title is misleading. Already long ago e.g. Kulmala et al. (2004) and Hirsikko et a., (2005) presented size dependent growth rates in sub 5 nm particles. The better title would be: "Size and time dependent growth rates of sub 5 nm particles".

See response to R1.1.

**R2.2** In my point of view it is missleading to present results in geometric diameter, since the calibrations of key instruments have been made using mobility diameters. Please, use mobility diameters in your text.

See response to R1.2.

**R2.3** In my view, the main novelty of the paper is the new method for determining the size-dependence of GR, which is now presented in Appendix A - but currently in a slightly confusing way. If I have understood the method correctly, the determination of Kappa(i) with the method presented is essentially a solution to a system of coupled equations of form A11, but with different Kappas at the upper and lower end of each size interval. And, the iterative procedure presented in the Appendix is a method to solve these equations. If this is the case, and, since this new method is an important part of the paper, I suggest writing these 'core equations' to the paper itself, and not having them in the Appendix (the description of the iterative solution can be left in the Appendix). Also it is crucial to write the equations in easy-to-understand way.

See response to R1.4.

**R2.4** I am somewhat surprised with the error estimates. In my point of view the errors particularly at sub 1.6 nm are much bigger than estimated. The counting efficiency of DEG-CPC is going down. The charging efficiency is unknown. How one is able to have so small error estimates?

It is true that both the detection and charging efficiency become increasingly uncertain below 2 nm, as discussed in Appendix C2.1. Since uncertainties in particle composition impact both the particle charging and detection efficiency, we considered instead the uncertainties in the *product* of the two efficiencies ( $f_{\text{detection}} \cdot f_{\text{charging}}$ ). A systematic uncertainty of ±50% in $f_{\text{detection}} \cdot f_{\text{charging}}$ at 1 nm was assumed to represent a "reasonable" level of uncertainty in that quantity which is by no means "small", as it covers nearly a factor of 3 in the magnitude of $f_{\text{detection}} \cdot f_{\text{charging}}$ at that size. The assumed systematic uncertainty also is constrained by current understanding of sulfuric acid driven particle formation, where a systematic uncertainty of ±50% in $f_{\text{detection}} \cdot f_{\text{charging}}$ allows for a range in inverted number concentrations of 1 nm particles that does not exceed the measured sulfuric acid concentration.

Also, an assumed systematic uncertainty of ±50% in $f_{\text{detection}} \cdot f_{\text{charging}}$ does not directly translate to an uncertainty of ±50% in the calculated growth rate values. This is due, in part, to a partial cancellation of correlated errors when calculating GR (see Appendix 2.3.2).

See response to R1.3 where further aspects of the effects of uncertainties in charging and detection efficiency on calculated quantities are discussed.

**R2.5** I am amazed how well the SMPS and Cluster CIMS data match (fig. A4). Is this typical (or some kind of 'best case' result)? Are some kind of fitting factors used?

See response to R1.5 where comparison of DEG SMPS and Cluster CIMS data is discussed.

**R2.6** Is the result in figure 2 a single measurement (a single time)? If yes, does the functional form of Kappa(Dp) vary a lot (at different times)? Would it be possible to plot some kind of a probability density plot of all measured Kappa-values?

The results in Figure 2a show growth rate enhancements $\Gamma\left(D_p\right)$ obtained from size distributions measured at two times (09:50 and 13:00) for a particular NPF event during NCCN. The results in Figure 2b are values of $\Gamma\left(D_p\right)$ obtained from 5 size distributions measured over a relatively short time period (12:40 – 13:00) during peak particle production for an NPF event during the NCAR campaign. Generally, during periods of nucleation, $\Gamma\left(D_p\right)$ is seen to increase with size for events measured during both campaigns. A probability density plot of all measured $\Gamma\left(D_p\right)$ (as a function of particle size and time) would be helpful in understanding the range of observed growth rate enhancements and potentially coming up with representative values for a particular environment/time of year. This is a task that we are endeavoring to accomplish in future studies.

**R2.7** Which of the presented methods, the steady-state one (Appendix A2.2) or the one where time-change dN/dt has been taken into account (Appendix A1.2), has been used to calculate the Kappa(Dp)-results?

Values of $GR_{OBS}\left(D_p\right)$ and $\Gamma\left(D_p\right)$ from NCCN events (plume events) were obtained using the steady-state method described in Appendix A2.2, while $GR_{OBS}\left(D_p\right)$ and $\Gamma\left(D_p\right)$ from NCAR events (regional events) were obtained using the method described in Appendix A1.2. Both methods are able to obtain these quantities, and determining which method to use depends upon whether the air mass that is being sampled is more regional or more plume-like in nature. The following text has been added in section 2.2 to make this distinction clear:

"The methods of analysis to obtain both $GR_{OBS}\left(D_p\right)$ and $\Gamma\left(D_p\right)$ for regional and plume events are described in Appendices 1.2 and 2.2, respectively."

**R2.8** It would be very good to compare and refer to some earlier papers namely Lehtinen et al. (2004) and Verheggen et al. (2006), which has very similar philosophy behind their growth rate analysis than present paper.

Citations to both mentioned studies have been added in the following amended text in section 2.2:

"Size and time-dependent observed particle growth rates, $GR_{OBS}$, were estimated using two new methods based on fitting measured size distributions to the aerosol general dynamic equation (GDE) (Gelbard and Seinfeld, 1978). Whether the measured size distributions were consistent with sampling from a regional air mass or with interception of a local plume determined the appropriate method to obtain growth rates from the measured size distributions. The analysis method for plume events utilizes the novel result that size distributions (< ~ 5 nm) for a nucleating system in the presence of an aerosol achieve pseudo steady-state shortly after the start of nucleation (McMurry, 1983). For regional events, the analysis method is similar in principle to earlier analysis techniques (Lehtinen et al., 2004; Verheggen and Mozurkewich, 2006) that fit size distributions to the GDE."

**R2.9** When comparing the contribution of sulphuric acid to the growth rate the newest results by Nieminen et al. should be mentioned and compared.

A citation to this study has been added in the introduction in the following text:

"Growth rates based solely on the condensation of sulfuric acid vapor significantly underestimate the observed growth rate (Sihto et al., 2006; Riipinen et al., 2007; Iida et al., 2008; Kuang et al., 2010; Nieminen et al., 2010) largely because organic compounds are responsible for up to 95% of the growth (Mäkelä et al., 2001; O'Dowd et al., 2002; Smith et al., 2008; Smith et al., 2010)."

**R2.10** The word first has been used too many times. It is proper in some cases, particularly when separation between time and size dependency is presented, but not in other times.

See response to R1.1.

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