Atmos. Chem. Phys. Discuss., 11, C10847–C10850, 2011
www.atmos-chem-phys-discuss.net/11/C10847/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "First size-dependent growth rate measurements of 1 to 5 nm freshly formed atmospheric nuclei" *by* C. Kuang et al.

Anonymous Referee #1

Received and published: 25 October 2011

General comment:

The article is well written and fits to the scope of the journal. It reports GRs in the size range that has only recently been covered by direct measurements. It presents an important contribution to the current knowledge of atmospheric new particle formation. I especially appreciate how the obtained results are brought to a larger perspective by using them in CCN survival probability calculations. I recommend publishing the article in ACP after the following comments/questions have been successfully answered.

Specific comments:

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). 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). 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). 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.

2) I cannot see any good reason to use geometric diameter of particles. As the DEG-SMPS 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 size ranges in figures should also be consistent (C2 and C3a have different axis than all others, and Figure A4 even uses mass diameters).

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). 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.

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.

5) How was the time periods in Fig 1 chosen? 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. How do the GRs compare to those calculated from the conventional SMPS? 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)?

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). Please give the numbers in the same order (Atlanta before Boulder) as they are usually discussed in the text. The sentence about enhancement factors (p. 25428, rows 13-19) is also very long and hard to follow, could you reformulate it?

References: Benson et al. (2011). Atmos. Chem. Phys., 11, 4755–4766. Hirsikko et al. (2005). Boreal Environ. Res., 10, 357–369. Lehtipalo et al. (2009). Atmos. Chem.

Phys., 9, 4177-4184. Lehtipalo et al. (2010). Atmos. Chem. Phys., 10, 7009-7016. Manninen et al. (2009). Atmos. Chem. Phys., 9, 4077-4089. Metzger et al. (2010). P. Natl. Acad. Sci. 107, 6646–6651. Sihto et al. (2006). Atmos. Chem. Phys. 6, 4079-4091. Sipilä et al. (2009) Aerosol Sci. Technol., 43, 126-135. Yli-Juuti et al. (2011). Atmos. Chem. Phys. Discuss., 11, 21267-21317.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 25427, 2011.