

Interactive comment on “Atmospheric new particle formation: real and apparent growth of neutral and charged particles” by J. Leppä et al.

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We would like to thank the referee for the constructive comments to our manuscript. Below are our answers to the comments. Major additions to the text are also quoted here.

The manuscript is generally well written and contains important and comprehensive new results. The derived analytic formulae will be useful for future growth rate analysis of new particle formation events in the atmosphere. I recommend the article to be published after my comments have been considered by the authors.

I can find several good motivations to why you have developed the analytical formulae to estimate the growth rates. However you don't clearly mention them in the paper. I

C1721

think you could advertise your article in a better way. What will you use the analytic formulae for in the future and why are they important?

1. The introduction is well written. However, partly I still miss the answer to why it is important to derive simple analytical formulae to estimate the growth rate of a nucleation mode due to self-coagulation and the apparent growth rate due to coagulation scavenging by larger particles. I think that one of the main motivations is that it is important to be able to clearly specify the condensation growth rate due to condensing vapors from measurements of the particle number size distribution in the atmosphere and not just the apparent growth rates. In section 4 you describe that this is important for correct estimates of the concentrations of condensable vapors. I think you should include this information in the introduction too, and clearly write that this is one of the motivations why you have derived the analytic formulas for self-coagulation and coagulation scavenging. Another important motivation could be that the analytic formulas can be used to estimate the importance of the charge enhancement on the growth rate in the atmosphere.

The motivations related to estimating the condensable vapor concentrations as well as demonstrating the effect of particle charges on the growth rates have been added to the introduction. The following part was added:

"The coagulation of particles can easily be simulated by detailed aerosol dynamical models, but retrieving the growth rate caused by the coagulation is not so straightforward. The particle growth rates observed in the measurements have been used to estimate the concentrations of condensable vapors often assuming that the growth is purely due to condensation (see e.g., Kulmala et al. 2001; Kristensson et al. 2008). Our aim is to provide simple methods to estimate the growth rate due to coagulation processes for the analysis of both simulated and measured data. Another possibly important, but very complicated, issue is the effect of particle charges on the particle growth rate. This topic is also covered in this study with the emphasis on the effect of charges on growth due to coagulation processes."

C1722

2. Section 2 "Tools and methods" was interesting to read and it is relatively easy to follow the derivation of the simplified analytic equations which are tested against the detailed aerosol dynamic model results. I have no other general comments on this section.

3. Figure 5. I don't understand why you use condensation sink as a proxy for the coagulation sink and not the actual coagulation sink. You write that you use the condensation sink as it is diameter independent. Don't you want to illustrate the diameter dependence of the coagulation scavenging which is caused by the fact that the coagulation sink depends on the diameter of the particles? Could you clarify this to me?

The condensation sink is used just to describe the amount of larger particles. It is used instead of coagulation sink to keep the Figure 5 readable, as the condensation sink has just one value, but the coagulation sink depends on the diameter of the particles. Using an average coagulation sink over given size ranges used in the figure could work, but it could also be misleading. Furthermore, the condensation sink is commonly used in the literature to describe the amount of aerosol particles, so the values are probably more familiar to the reader than values of coagulation sink would be. To make this clearer, the wording is now changed to the following:

"Here, the condensation sink, rather than coagulation sink, is used to describe the amount of pre-existing larger particles in order to simplify the figure, as the condensation sink has just one value, but the coagulation sink depends on the size of the particles."

4. Section 3.3.3 "Semi-apparent growth due to self-coagulation". First I did not understand that by self-coagulation you mean the coagulation between all the particle sizes within the nucleation mode and not just the coagulation between particles within the same size bin. If self-coagulation would be considered only between particles with exactly the same size (same size bin) there would not be any semi-apparent growth due to self-coagulation. Please clearly state what you exactly mean with self-coagulation

C1723

in the beginning of the text. I did not find this information.

The used definition of self-coagulation is now added to the Section 2.1.1. Also the applicability of Eqs. 8 and 9 (growth rate due to self-coagulation) is stated in the end of Section 2.1.3. With these clarifications, the concept of semi-apparent growth due to self-coagulation should be clear as it is written in Section 3.3.3.

5. Section 3.4 "Charges". Interesting results and discussion but it is much information and you need considerable amount of time to understand what Fig 6 shows. Are the red dashed lines in Fig 6 displaying the atmospheric most realistic conditions? It could be good to give this information. That all formed particles should be charged (50/50) seems unrealistic for atmospheric conditions, or am I wrong? Can the fractions of charged particles change significantly from time to time and from one location to another, and could this then explain observed variability of the growth rate? I understand that these questions cannot be answered within the scope of this article but it would be interesting if you could address these questions in the future. Since the analytic formulae provide the tools to answer these questions you could mention this as a motivation of why you derived these formulas.

In the literature, there are quite many studies suggesting that the percentage of ion-induced nucleation (IIN) in the atmosphere varies within 0-40 % depending on the measurement conditions, but even values as high as 100 % has been suggested. In this study, we do not intend to emphasize our educated guess on the subject. For this reason, we cover also the extreme possibility of 100 % IIN, which also gives us the maximum influence that the particle charges can have on the growth rate. This reasoning is mentioned in Section 2.3.3 where we describe the parameters varied in the simulations.

6. I understand that the Ion-UHMA model simulations are important to test the simple analytic formulas for self-coagulation and coagulation scavenging, since this information cannot be derived from measurements. However, otherwise I don't really see why

C1724

the simplified analytic formulas should be applied to model simulations which already contain the information about the concentrations of condensable vapors, coagulation sink etc. Hence, I don't know if the text on line 19-27 on page 2105 is relevant or not. Do you mean that one important application of the simplified analytic formulas is to address the influence of the fixed sectional approach and the numerical diffusion on the model results? Then I think this should be clearly stated here. For complex conditions observed in the atmosphere with influence from coagulation, condensation and new particle formation at the same time I can understand that this method could be valuable to test the model performance, concerning the numerical diffusion. I thought that the model simulations were used mainly to test the analytic formulae and that the main application of the analytic formulae is for measurement data. If this is the case I think you should reformulate the last sentence in the abstract.

Indeed, the vapor concentrations are usually known when conducting aerosol dynamical simulations. The reason to mention the simulations in Section 4 is to remind of the error in the condensational growth rate, which might be important when methods used to determine the concentrations of condensing vapors are tested on simulated data. The error caused by the numerical issues is discussed in more detail in Section 3.1, but as they have implications on growth rate analysis related to simulations, the following paragraph was added to the Section 4 as a reminder:

"In aerosol dynamical simulations, the concentrations of condensing vapors are usually known. Thus the difference between the real and the effective condensational growth rate can be used to estimate the error in the condensational growth rate due to numerical diffusion and the division of particles into sections, when known concentrations of condensing vapors are used. This underestimation can easily be of the order of 20 %, but at least a part of this error can be taken into account using Eq. 24 to calculate the effective growth rate."

One of the purposes of this study is to provide means to determine accurate growth rates from sectional simulations. Though the coagulation rates in the models are very

C1725

well known, it is not so straight forward to determine the growth rate due to coagulation, which is a property based on the changes in the population. As a second application, these methods can be used to determine the growth rate due to coagulation from the measurements.

Specific comments: Line 10 in abstract. Try to avoid to use the phrase "work quite well" since this information is subjective.

The phrase "work quite well" is now changed to "give accurate values".

Page 2083, line 2-11. I think you should clearly write that the constant growth rate you talk about is for non-volatile compounds only. I assume that the reason why you see an increase in the condensational growth rate as a function of particle diameter from measurements is that the much of the condensing material is semi-volatile.

The non-volatility assumption of condensing vapors is stated earlier in the Section 2.1.2. To avoid misunderstandings, it is now mentioned again when stating that we use the assumption of constant growth rate in this study.

Figure text to Fig. 6. A fraction cannot be larger than 1. Either you should change the legend of the figure or in the text to percent instead of fraction.

Word "fraction" was changed to "percent".

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 2077, 2011.

C1726