

Interactive comment on “A numerical comparison of different methods for determining the particle formation rate” by H. Vuollekoski et al.

Anonymous Referee #1

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Overview

In this article the author uses the aerosol dynamic model UHMA to investigate the determination of formation rate using different methods. The authors conclude that estimates of formation rate based on measured number concentration N3-6 agrees well with more detailed calculations of the formation rate. The authors further conclude that the widely adopted power-law dependence between nucleating vapors and formation rate is inaccurate.

The subject is within the scope of ACP, and most of the time a proper language is used. However, in its current form, it is occasionally hard to follow the discussion and arguments. Re-writing some parts would increase the accessibility of the paper.

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Furthermore, the authors make some, in my opinion, unsupported statements that needs further treatment and discussion before the article could be published in ACP.

Major comments

In the introduction part, the author state that using a simplified model has advantages compared to analysis of measured data. This is due to the fact that measurement data is associated with numerous processes that interfere with the analysis, such as changes in air masses and boundary layer height. Then however, the authors still conclude that the derived results are applicable to analysis of measurement data. Can the author expand on this? How would the knowledge gained through this study be transferred into more complex systems where the omitted processes *de facto* are active? For example, can this statement be justified when the authors adopt highly simplified assumptions regarding the nature of the organic compounds participating in the initial steps of the growth (i.e. I assume it is single low volatile compound)? The nature of this compound (thermodynamic and molecular properties) is furthermore not discussed in the MS.

As a main conclusion, the authors state that the often used power law dependence is inaccurate. As the MS is written, I do not see any clear support for this statement. The power law relation is used to connect the concentration of nucleating material ($[H_2SO_4]$) to apparent formation rate. The author's base this conclusion on the fact that maximum in sulfuric acid concentration not necessarily coincide with maximum in J3. This statement is rather strong, and requires more attention in the MS. The authors should provide tentative explanations for this mismatch. This statement should be backed up by analysis of when and why the power law fails, and when and why it is applicable. What is the role of the initial cluster size? What is the role of the nature of the organic component? What is the role of the background aerosol? How big fraction of the mismatch can be attributed to errors in calculating the time shift? The authors seem to suggest numerical diffusion to be a reason for this mismatch; if so, what would be the implications for measurement data?

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Specific comments:

Page 18792: "The effects of numerical diffusion were not considered in this study." Can you comment on the expected importance of numerical diffusion and what factors that would influence the error induced by it?

Page 18788, equation 5 and related discussion: Can the author explain why GR around 3-7 nm is used?

Page 18791: Equation 8 is a suggested improvement over the analysis of J3. It is however not clear to me if this improvement is the result of trial and error, or if it is based on physical considerations. The text is a little bit unclear in this perspective. Just tuning in the equation using different size ranges does not have any real implications for treatment of measurement data. How will these assumptions hold if the resolution of the instrument is changed, as example?

Page 18791, line 20: "...most of the deviation between Eqs. (3) and (5) occurs because of coagulation...". What do the authors mean by this, is it that the calculation of coagulation is wrong? Try changing to "...is caused by..." or "...occurs due to error in the estimate of the coagulation term..." Furthermore, doesn't the result in table 1 (BKG→0) indicate that coagulation is completely responsible for the deviations? Or is it also connected to the CS? Please comment on this.

Page 18792, line 21-22: Please clarify what you mean here.

Page 18792, line 6-7: Do not get what the authors mean by phenomena here. Do we mean processes, size ranges or what?

Minor comments:

Page 18788, line 5: "...basing on them..." → "...based on..."

Page 18789, line 17-18: " For an example..." → "As an example..."; "For example..."

Page 18789, line 18: Can a background be "more abundant"? Sounds awkward. In

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fact, the whole sentence needs rephrasing as it is currently hard to understand what the authors really mean.

Page 18790, line 7: "We modified the analysis to be more accurate..." sounds a little bit subjective. Maybe should write "We performed more detailed calculations..."

Page 18790: Line 10: "...comparing these...", should be stated more clearly what you are comparing here in order to improve readability.

Page 18791, line 1-5: Please clarify.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 18781, 2010.

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